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Applied

# NUCLEAR PHYSICS

SECOND EDITION

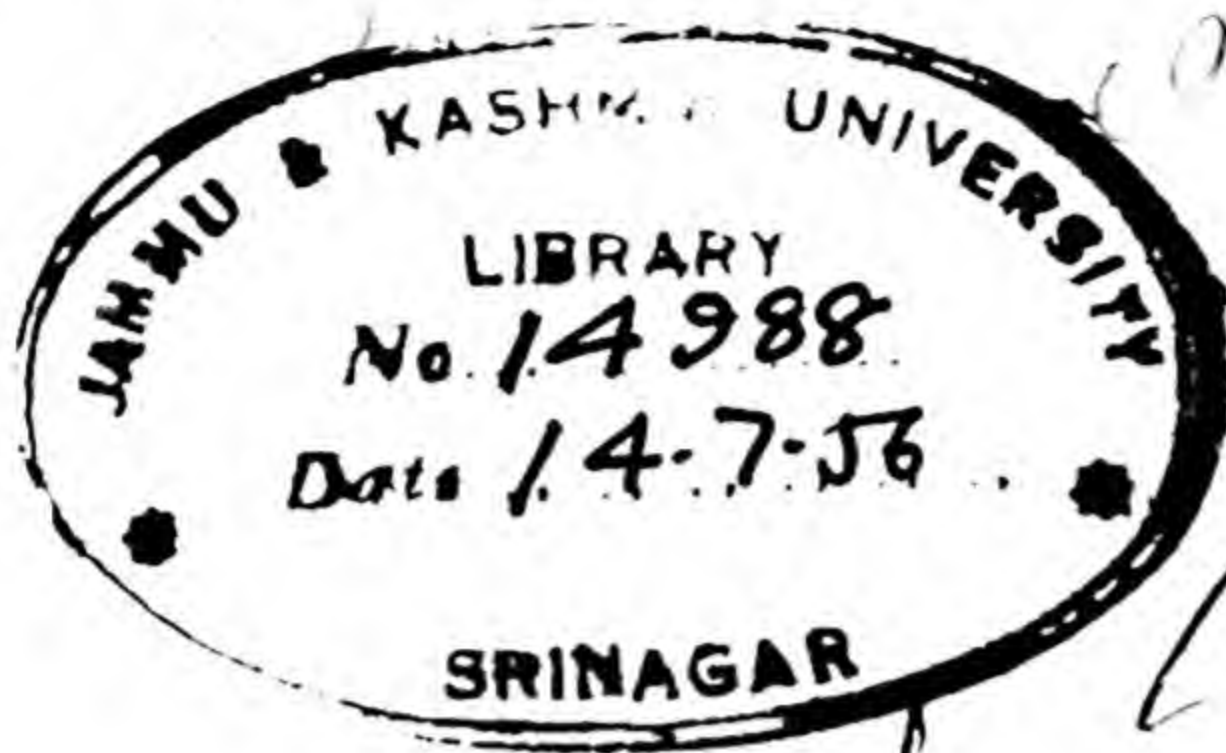
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JOHN WILEY & SONS, INC. · NEW YORK  
CHAPMAN & HALL, LTD. · LONDON



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SECOND EDITION

*Second Printing, November, 1953*

Printed in the United States of America

539.7





## Preface

The first edition of this book appeared in 1942, during the early days of World War II. As we look backward, the phrase "applied nuclear physics" seems more of a chauvinistic hope than an accomplished reality. Yet, even as the pages came from the presses, Fermi and his cohorts were busy in Chicago fashioning that historic structure of graphite and uranium which was to herald the birth of the atomic age and broaden tremendously the potential applications of nuclear science. Thus the same act that gave muscle and sinew to applied nuclear physics—the concept—signaled obsolescence for our book bearing the same name.

The present edition represents an effort on our part to remedy this situation. A new chapter on nuclear chain reactions has been added. Special sections have been devoted to pile theory, neutron diffraction, cross sections, and cosmic rays. Also included for the first time are detailed instructions to guide the reader who may wish to perform a number of informative laboratory experiments in nuclear physics. The tables of nuclear data have been brought up to date. Much of the subject matter common to both editions has been rewritten and expanded in the light of advances of the past eight years. In a field of science receiving as much attention as is being accorded nuclear physics today it is inevitable that the tempo of progress will be rapid and ever forward. However, it is our modest hope that the ensuing pages give a reasonably balanced and accurate picture of nuclear science and technology as they exist today—1950.

The reader may question the preceding statement in view of the absence of discussions relating to atomic weapons. Atomic bombs, thermonuclear reactions leading to an hydrogen bomb, and radiological warfare are scarcely mentioned. Two reasons account for these omissions. In the first place, security regulations would prevent our discussing these subjects in other than general, speculative, and watered-down terms. A second and more insistent rationale prompts us to omit these items. Thus far, the utilization of atomic energy for



destructive purposes has had an impact on civilization far outweighing the beneficial gains. And as we stand today, perhaps in the shadow of another world catastrophe, it requires a brave man indeed to suggest that the scales will be balanced tomorrow or even a century hence. Yet it is impossible for us to believe that the ultimate destiny of this primordial force is to be one of devastation, desolation, and chaos. If this be so, then, truly, the efforts of that band of scientists, laboring patiently over the years to wrest these secrets from nature, have been in vain. Rather must we believe that in the final analysis it is the constructive, good, and useful side of this double-edged sword which will prevail. Hence it is these which are stressed in this book.

No subject in science has been studied with as much sustained interest as the transmutation of the elements. Transmutation is essentially linked with the atomic nucleus, and its study will continue to be of utmost value in elucidating the characteristics of this smallest domain of nature. Undoubtedly this study will yield new and important theoretical ideas. Yet it is doubtful that these ideas ever will have the influence in fields outside the realm of physics that is already exerted by the technical developments in nuclear physics. The discovery of artificial radioactivity has put in the hands of chemists, biologists, engineers, and medical workers the means for studying individual "tagged" atoms, which permits experiments that could not have been contemplated a short time ago. The advent of nuclear reactors has parted the curtain on untold vistas in science, providing the physician with potent tools for the treatment of disease, the crystallographer with a new probe for deducing atomic arrangements in matter, the industrial scientist with unbelievable radiation fluxes which can serve to exert profound changes on metals and organic compounds, and an energy-hungry world with a source of concentrated power, possibly comparable in extent to our present known reserves of fossil fuels.

The technical aspect of nuclear physics is emphasized in this book. We aim at presenting the essential facts in such a way as to be of service to the growing army of scientists and engineers, who, though not necessarily versed in the language of physics, are using the products of nuclear physics in their respective spheres.

Although the elaborate theoretical approach to nuclear processes has been eliminated, we feel that the account is sufficiently thorough to make this a useful textbook in a course on nuclear physics.

We have not aimed at making this primarily a reference book, but rather one to be read for description and explanation. We have,

therefore, not compiled a complete table of references, which indeed would necessitate a much larger work than this, but we have provided a small number of references which can be consulted for more detailed information.

ERNEST C. POLLARD  
WILLIAM L. DAVIDSON

*January 1951*



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*Minnie*

# 1. Randomness, Reason, and Atomic Energy

The world is made up of atoms. These atoms are invisible, and indeed modern physics is in a sense founded on the belief that atoms can never be seen. Yet it is known that there are only a certain number of types of atom and that these types have certain quite definite structures which clearly explain the properties of the atoms. Atoms are tiny—about  $10^{-8}$  cm across, and they consist of electrons rotating about something much smaller yet, the atomic nucleus, which is about  $10^{-12}$  cm across. The nucleus, which is so tiny, is the storehouse of energy in the universe, and recently that storehouse has been tapped by mankind, for what is perhaps the first time.

This book is concerned with the nucleus of the atom, a realm of nature completely foreign to our senses, forever far too small to be seen. Its properties are largely unknown, and it offers a challenge to scientific research which is perhaps unequalled by any other object of study. Yet an impressive number of facts are known about these invisible nuclei, and soon these facts will be described. The authors have told this kind of story many times, and invariably the first reaction of the audience is the question: "How do you *know* these things about atoms and nuclei?" Therefore first we propose to spend a part of this chapter indicating how this knowledge is gained.

The method used involves faith in a theory which predicts the results of experiments accurately. The first such theory of interest here was that of Dalton who proposed that chemical elements could be divided into smaller and smaller units, just so far and no more. That limit was the atom of the element. The prediction made from this was that elements would combine with one another in definite proportions by weight, and not in variable proportions as in a solution. Careful experiment showed that for a wide variety of combinations this prediction worked, and the atomic theory accordingly was accepted as a basis for explaining chemical combinations. The second theory was



the kinetic theory which proposed that matter, particularly gases, is made up of molecules in rapid motion and that the effects of such motion include temperature and pressure. This theory required that the number of molecules in even small volumes be enormous. The conclusions drawn were by no means so clean-cut as those of the original hypothesis of Dalton, for the reason that complete predictions would have to involve the structure of the individual atoms, an unknown factor. Nevertheless the theory had some confirmation, such as the verification of the prediction that the viscosity of a gas does not depend on the pressure for wide pressure ranges.

The first real success of this theory came when means for producing nearly evacuated spaces became available. The electric discharge became a phenomenon which could be studied accurately, and in particular it was found that, when pressures were finally obtained low enough to permit molecules to traverse a tube without collision, charged particles called electrons were emitted from the cathode. These electrons were part of atoms, electricity was apparently atomic in origin, and a wave of new experiment and theory began.

The first prediction based on the new atomic theory was that all electrons are alike. This meant that they should all have the same charge, mass, and any other property of importance. The fact that they had the same charge was proved by J. J. Thomson, C. T. R. Wilson, and most convincingly (though later) by R. A. Millikan. With this proof went the consequence that all electrons had the same mass, since the deflection in a magnetic field depends primarily on the ratio of charge to mass and this had already been shown to be a constant.

The next prediction brought a great upheaval in thought, which is only now subsiding. This was that the atom consists of a solar system with electrons for planets and something unknown for a sun. The unknown was called the "nucleus." Since the electron had been shown to be light, the nucleus had to be heavy. Since the electron is negative and matter is neutral ordinarily, the nucleus had to be positive. Since the electron had a fixed charge, the nucleus had to have multiples of this fixed charge. All this is quite sensible. The nasty point concerned the fact that all the electrons in the atom rotate about the nucleus, they are therefore accelerated, and therefore they should do what any other accelerated charge should do—they should radiate electromagnetic energy. This energy they accordingly should lose, and in losing it they should fall toward the nucleus. The birth of such an atom would therefore also be its death. This inescapable



conclusion meant that the birth of such a theory was also its death. A theory, however, lives in the minds of people. It died for most scientists, but not for Rutherford. Rutherford had turned his experimental skill on the subject of radioactivity and with a tremendous instinct had fastened on the "alpha" particle, known to be emitted from many radioactive elements. This particle is easily stopped, yet it tears the electrons from hundreds of thousands of atoms before it stops. It does this so effectively that single particles produce feeble but observable scintillations in zinc sulphide crystals.

Rutherford proved indirectly and directly (by collecting enough particles to produce a spectrum) that these particles are helium atoms in some form or other. He proved that they are doubly charged, and that their mass is nearly that of the helium atom itself. He began to use these helium particles to bombard matter. The results of his work resuscitated the nuclear theory of atoms, and today it is firmly established. To use it, however, we require wholly new concepts of motion and of radiation.

The process of atomic bombardment is one of great interest. The alpha particles are produced at random from the radioactive element used as a "source." The atoms in a gas or even a solid target must be considered as distributed at random. One's first reaction is that this combination of randomness can result only in complete confusion. Yet it doesn't. Firm and sure conclusions can be drawn from such experiments as Rutherford's.

The reason is that the large numbers of atoms in materials offer a statistician's paradise. Averages can be taken over billions of billions, not a few tens. With such averages great precision can be attained. It is this fact which makes a science out of atomic bombardment.

Consider a set of gold atoms in a foil. There are, say  $10^{17}$  of them. Now suppose that each has an area of  $10^{-16}$  sq cm, as would be expected from an atom with a diameter of approximately  $10^{-8}$  cm. The total area the atoms present is therefore actually 10 sq cm. If the area of the foil happens to be less than this, then an atom fired at the foil should bounce and not go through. Rutherford knew that, although a few alpha particles bounce, most go through, and through much thicker foils too. He therefore argued that the atoms must be smaller, certainly from the point of view of an alpha particle. If it could be argued that alpha particles are helium *nuclei*, far smaller than helium atoms, and that they are not affected appreciably except by other atomic nuclei, then the transparency of the gold can be ex-



plained. Rutherford therefore took this as a hypothesis. He suggested that, if the alpha particles which actually do bounce from the gold were carefully observed, they would prove to be distributed as if they had been charged particles deflected by the field of a gold nucleus. Rutherford mastered the mathematics of this kind of deflection and found that the number scattered through any angle should be proportional to the fourth power of the cosecant of half that angle, and also that at any angle the number scattered should depend on the inverse square of the energy of the particle. Geiger and Marsden showed that these two queer conclusions were valid. They showed further that the nuclear charge required for helium and gold were those required by the position of the two elements in the periodic table. Certainly Rutherford was convinced of the reality of the nuclear atom.

Then came one of the most curious twists that has ever happened in science. The nuclear atom was used by Bohr to explain the spectrum of hydrogen, a spectrum well known to be simple, but with a baffling simplicity that fitted nothing else in physics. To be understandable in 1913, spectra had to be like the overtones of an organ pipe, with frequencies in multiples of whole numbers. The hydrogen spectrum had frequencies dependent on whole numbers, but inversely on the square of whole numbers. Bohr showed that precisely the observed relation could be expected with the nuclear atom, provided one started with some new postulates. In this Bohr showed the imagination necessary to comprehend the new and invisible part of nature. *Nature is built up from atoms, not down to atoms*, and clear understanding of nature can be gained only by starting from atoms and thinking outward, not by attempting to conceive of atoms as small-scale reproductions of what we are used to seeing. Bohr perceived this and by his postulates founded the first valid atomic theory. Today these postulates are part of a more complete theory, quantum mechanics, and appear as necessary consequences of the theory. Nevertheless Bohr's theory made it finally necessary to believe the atom to have a nucleus, small and charged in units of the electronic charge, with a positive sign and a number of units characteristic of the chemical properties of the element.

It is interesting to see the divergence in interest of the two great men, Bohr and Rutherford. Bohr followed keenly the intense development of the understanding of the electrons which go in orbits around the nucleus, a development which went furiously forward in every



nation. Rutherford stayed resolutely with the nucleus he had discovered.

To do this required much imagination. The tiny nucleus is the least theatrical part of the atom, and it took much insight to believe that it played a dominant role. Yet Rutherford, with the belief that radioactivity originated in the nucleus and with an intense interest in radioactivity, was convinced that the fireworks were in the nucleus. He held this conviction even though the excitements of the chemist and spectroscopist were all concerned with the outer part of the atom. Later events have proved him right.

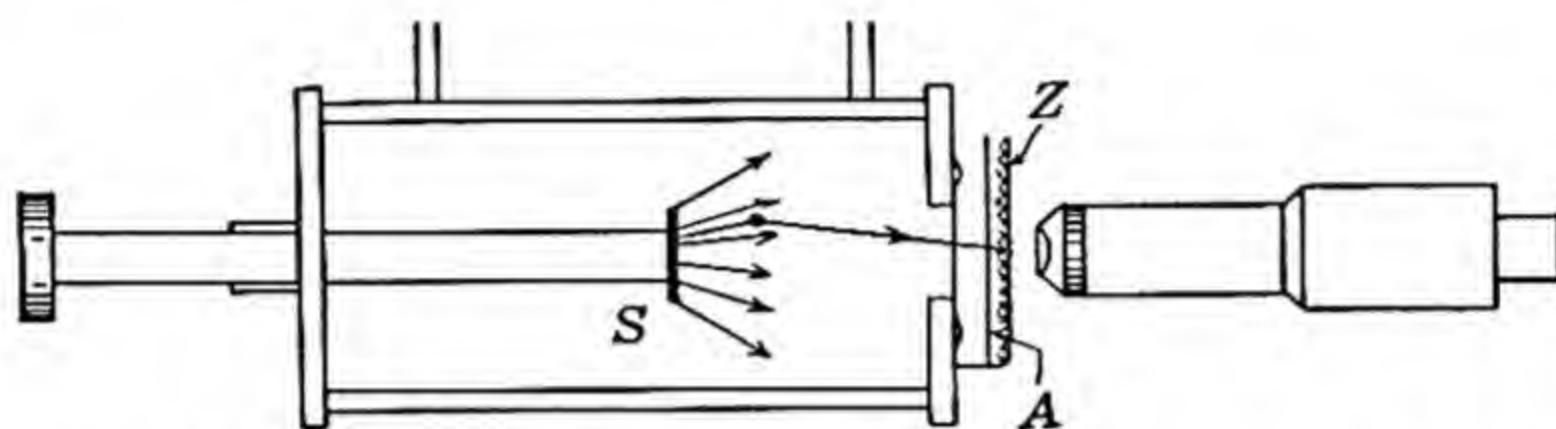


FIG. 1. Rutherford's original apparatus for the transmutation of nitrogen into oxygen. Alpha particles from the source bombard nuclei of nitrogen and cause the transmutation. At the same time swiftly moving hydrogen nuclei are set free, which traverse a considerable thickness of absorber and are detected on the scintillation screen.

Rutherford therefore sought stronger and stronger sources of alpha particles. With these sources, one of his students, Marsden, found that hydrogen when bombarded by alpha particles gave rise to new particles of longer range and somewhat different character. Rutherford proved that these were hydrogen nuclei, or protons. And then, in the strained and tense atmosphere of World War I, Rutherford began the experiments that have led directly to the release and partial control of atomic energy. Rutherford found that these protons were produced from air as well as from hydrogen. However, air has to be dried carefully lest there be hydrogen nuclei in water vapor. The careful proof that protons can be knocked out of nitrogen nuclei by alpha particle bombardment had to wait until 1919. Then Rutherford was able to show that not only were the protons produced far too abundant to be caused by traces of hydrogen but that they also had more energy than ever before observed. These protons were hydrogen atoms which had been produced from a totally different element, nitrogen. The apparatus for this, the first transmutation ever produced by man, is shown in Fig. 1. It is in striking contrast to the elaborate equipment of modern atom smashing.



$S$  is the source of alpha particles (a deposit of  $\text{Ra[B + C]}$  on a metal disk);  $Z$ , the particle detector, a zinc sulphide screen which emits faint scintillations when single alpha particles or protons strike it;  $A$ , a series of absorption screens which consist of thin mica or aluminum and can be placed between the gas in the box and the zinc sulphide screen. When hydrogen was introduced into the box, as in

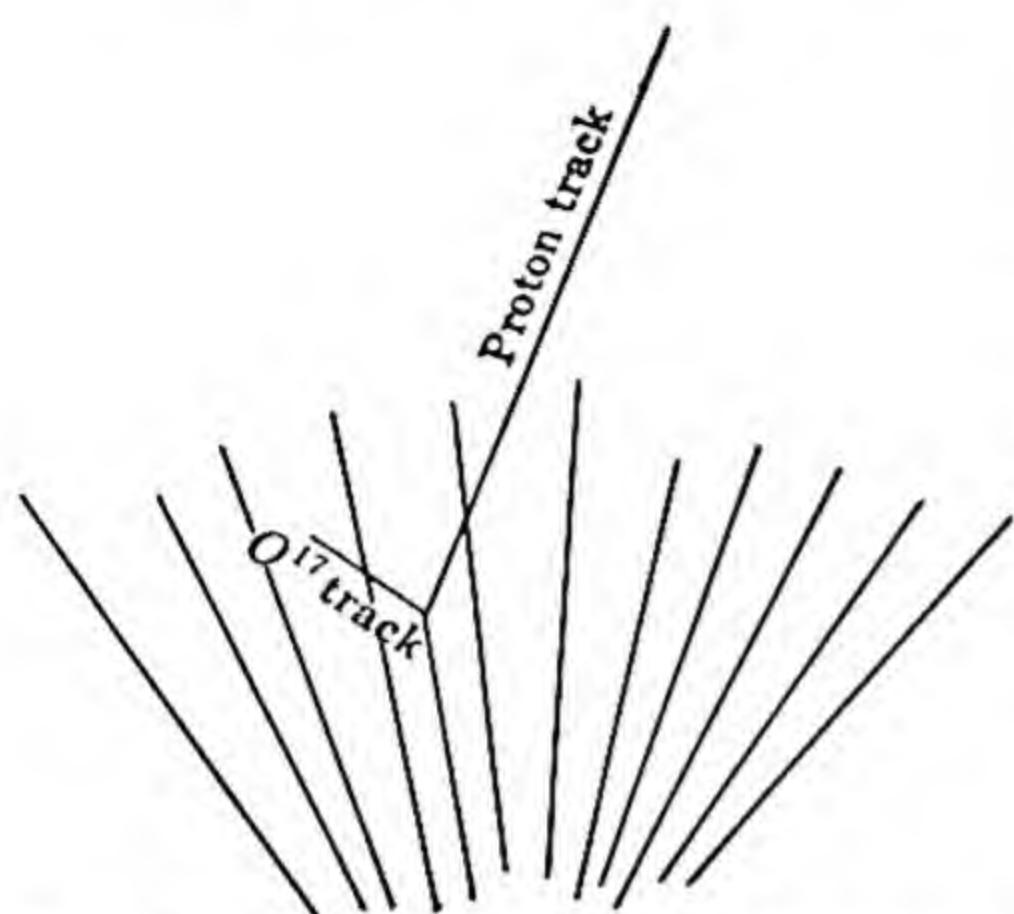


FIG. 2. Diagrammatic representation of a cloud-chamber picture showing the occurrence of a transmutation event. Of the numbers of wasted alpha-particle tracks, one is seen where the alpha particle has combined with a nitrogen nucleus with the emission of a proton. That this combination takes place is shown by the absence of a fourth track due to the spent alpha particle.

the experiments of Marsden, it was found necessary to introduce about four times the thickness of absorber to stop all scintillations from reaching the screen. This increased thickness is easily explained, for in the bat-and-ball kind of collision the alpha particle acts as the bat and is four times heavier than the hydrogen nucleus, the ball. Therefore an increased penetration by the lighter hydrogen nucleus, or proton, is to be expected. However, beyond this absorption no scintillations at all were found. When carefully dried air was introduced into the box, a few scintillations were found beyond this point. Rutherford correctly concluded that the nucleus of nitrogen had been in some way invaded by the alpha particles and that new particles were produced.

The nature of this transformation was not at first understood. It might

be a shattering of the nitrogen nucleus into fragments; it might be a detachment of one part of the nucleus; or it might be a kind of chemical reaction in which helium and nitrogen combined and new elements were formed. The nature of the process was finally conclusively established by the experiments of Blackett in the Cavendish Laboratory and independently by Harkins at Chicago. A representation of a picture taken by Blackett is shown in Fig. 2. Blackett and Harkins independently undertook the laborious procedure of photographing many times a small number of alpha-particle tracks in a Wilson cloud chamber (which renders the paths of ionizing particles visible by the presence of droplets of water where the ions are formed), in the hope that one photograph would show a transmutation with the



expected thin long track of the ejected hydrogen nucleus. Such an event might be a grand catastrophe with many tracks visible, or a set of *four* tracks, one for the new hydrogen particle, one for the recoiling nucleus, one for the incident alpha particle, and one for the alpha particle afterward; or it might be a set of *three* tracks only, the alpha particle being absorbed with a new nucleus formed. It can be seen from Fig. 2 that there are only three tracks, those of the incident particle, the new nucleus, and the hydrogen particle or proton. The helium and nitrogen combine in a nuclear reaction to form hydrogen and a new nucleus.

Now a little reasoning about this process shows us something about structure in the nucleus. Helium of mass 4 is joined to nitrogen of mass 14, and hydrogen of mass 1 is taken out. Helium has charge 2, nitrogen 7, and oxygen 8. Suppose, then, that the basic cause of nuclear charge is the content of protons, a simple suggestion. If true, however, *there must be something in addition*, for the nuclear mass is not 7 for nitrogen and 8 for oxygen but double those numbers or possibly even more. So this first experiment in transmutation, which tells us that there are protons in the nucleus, also requires more than protons. This added part was surmised by Rutherford and discovered by Chadwick in 1932 to be neutral particles, or *neutrons*, of mass very nearly the same as the mass of protons, but having no charge.

The discovery of the neutron is a masterpiece of close reasoning combined with crucial experiment. It was discovered by Bothe and Becker in 1931 that the element beryllium when bombarded by alpha particles emitted penetrating radiation. Irene Curie and Frederic Joliot discovered that this radiation would interact with nuclei such as hydrogen and helium or even argon in a manner that required exceedingly high energy for radiation of the character of x-rays. Since such high energy radiation was being revealed in cosmic radiation, it did not seem out of the question that the newly transmutable nucleus could be emitting such cosmic rays. The catch was the fact that the energy as measured by hydrogen interactions was not too large, whereas the same radiation when interacting with argon seemed to be much more energetic.

This difficulty was resolved by Chadwick who proposed that the penetrating properties of the new radiation were due not to its being like an x-ray, but to the fact that it consisted of neutral particles moving in a world of intense electric charge but wholly unaffected by the existence of charge. The penetration was due to the fact that the only influence felt by a neutral particle is that of the nucleus, and



this is so tiny an object that collisions with it are very rare. Chadwick showed that the neutron hypothesis gave the same energy for interactions with hydrogen, helium, and argon; he then went further to the crucial experiment. If neutrons are emitted in the same way as protons, then the energy of the neutrons depends on whether they are emitted in the same direction as the incident alpha particle or opposite to it. If they are emitted in the same direction, the momentum of the alpha particle will add to the momentum of the neutron; if opposite, there will be a reduction. Moreover, the exact value of the two energies can be calculated by quite ordinary billiard-ball type of calculations. Therefore the energy of the hydrogen recoils should be definitely different, according to whether the neutron is going with or against the alpha particles. What is more, the energy values should agree with calculated values. Chadwick found that they did; and he proposed the existence of a neutron, a profoundly important discovery.

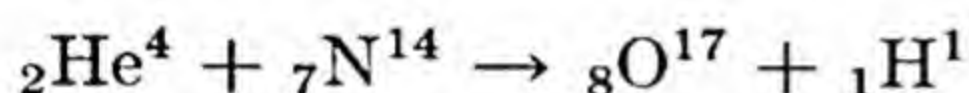
It has turned out that the three important building blocks of nature are the three elementary particles already mentioned: the proton, the neutron, and the electron. The proton is positively charged with the elementary unit of electric charge and has very nearly the mass of the hydrogen atom, namely,  $1.6 \times 10^{-24}$  gram. The neutron has practically the same mass but no charge at all. The electron has a negative unit charge and a small mass of approximately  $9 \times 10^{-28}$  gram. It is almost certain that these three particles are themselves special forms of a fundamental matrix of nature which we have not yet discovered. The use of high energy bombardment is showing that the nature of these three particles is considerably more complex than it seemed ten years ago. When the whole story is told it is likely that the complexity will yield to simplicity as it always has so far, but this assertion is not backed by proof as yet. In any event, for the very great majority of purposes, we can think of all matter as composed of these three particles.

The particles alone are not sufficient: there must be forces. The most familiar force, that between charged particles, the coulomb force, is well known. It is responsible for the whole of the external, i.e., chemical properties of the atom, simply on account of the force between protons in the nucleus and electrons outside. This force causes the electrons to rotate in stable orbits as previously described by Bohr, and the nature of the orbits of these electrons is responsible for the chemical behavior of the atom. The other forces at work between elementary particles are not nearly understood. They are exceedingly powerful, operate only over very small distances, and do



not appear simple for the adequate reason that as yet we do not know their cause. These forces operate between all nuclear particles, nearly, but not quite, equally. Since they are so potent, small differences in their nature have quite important consequences, as will be seen in a moment.

With these three building blocks the nature of Rutherford's pioneer experiment can be seen from Fig. 3. The nucleus of nitrogen is indicated as having seven protons and seven neutrons. The incident alpha particle is shown, and the result is the oxygen nucleus, or *isotope*, having eight protons and nine neutrons, with the proton detached. Such a process can be written as a kind of chemical reaction.



The subscripts refer to the nuclear charge or number of protons; the superscripts refer to the nuclear mass or sum of neutrons and protons. Since the symbol

for the chemical element really tells us the nuclear charge, subscripts are usually omitted. However, at first it is helpful to keep them in so that the reaction can be clearly comprehended. Note that the form of oxygen is unusual. The great majority of oxygen nuclei have eight protons and eight neutrons. However, some exist with nine and ten neutrons.

Now for a short while we need to shift our attention to cosmology, the science of the nature of the universe. If this remarkable atomic nucleus is built up of neutrons and protons and they can attract one another to form stable combinations, what combinations will be found? There exists definite evidence that the universe started its present phase some three billion years ago and that when it was young the concentration of energy was great. In this grand free-for-all stage the average energy of each particle was enormous, and all kinds of combinations could be formed and broken. We can imagine therefore that every possible nuclear species was present at one time. On the other hand, at this late date, we have a strictly limited number of nuclei, at least here on earth. There must, therefore, be some inherent reason why some nuclei are more stable than others. If we

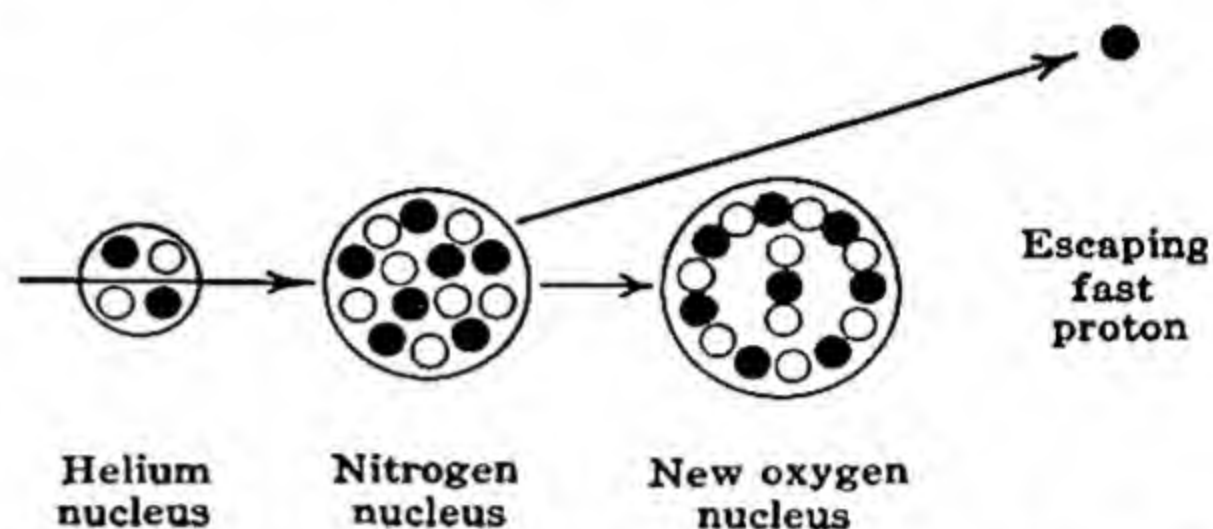


FIG. 3. Schematic representation of Rutherford's original transmutation. The helium nucleus consisting of two protons and two neutrons combines with the nitrogen nucleus consisting of seven protons and seven neutrons to form a rare isotope of oxygen consisting of eight protons and nine neutrons. A fast single proton is also emitted.



think about it a bit we can see one simple reason which must operate. The forces between protons are a composite of attraction, due to the true nuclear forces, and repulsion, due to the coulomb forces. If we load a nucleus unduly with protons, the repulsion will begin to act unfavorably, and a nucleus of the same number of particles but fewer protons is more stable. This simple reason is certainly not all. Just as in the outer electronic structure of the atom there are rules which determine the difference between elements, so in the nucleus there must be some set of principles operating to make some nuclear combinations strong and others weak. In the outer atom we know the structural rules: so far we do not know the principles operating in the nucleus. However, we find quite definitely that rules do operate and that there are more stable combinations and less stable ones.

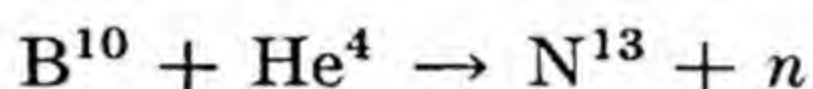
The short paragraph above is the key to understanding two big branches of nuclear physics: the release of nuclear energy, and artificial radioactivity. The question arises as to whether the process of evolution from the early energetic times to the modern stable times has gone uniformly, or whether some combinations of neutrons and protons are present today which could be induced to change to others which are more stable. If the latter, then the excess of energy which has been preserved from primordial times can be used by us. If this energy can be utilized to change still more atoms to more stable forms, then we have found a process by which we can develop the sealed-in energy continuously until the supply of material which has not fully evolved has gone. Uranium and thorium represent such materials from which we can derive nuclear energy. This was achieved in the Chicago chain-reacting pile in 1942.

The process of radioactivity is the last and feeblest method of nuclear evolution. If a really wild combination is made, such as silicon with fourteen protons and no neutrons, it will probably just blow apart into several smaller units. If a tolerably wild form is made, such as magnesium of mass 28, it will possess so much excess energy that its first action will be either of two things. Either it will split apart into smaller units, like two carbons of mass 14, or it will emit energy as radiation, leaving magnesium of mass 28 with no energy other than that appropriate to its nature. Now in either of these events, there is still another evolutionary process available. This process reveals the fact that the neutron and proton have some further structural feature, for it is possible for a neutron to convert to a proton, or vice versa, with the emission of an electron or a positive electron as required. This act of conversion can change nuclear constitu-



tion, so to speak, within itself. Thus carbon of mass 14 can become nitrogen of the same mass by changing a neutron to a proton and emitting an electron. Magnesium of mass 28 has a rather more vigorous change to make but can do so in two stages, first by emitting an electron and converting to aluminum of mass 28, and then by emitting a second electron to reach silicon of mass 28, which is stable.

It may be expected that, if the process of evolution from early times has not been uniform, then perhaps some nuclei have been left which are still in the process of reaching ultimate stability and have accordingly not yet changed neutrons into protons. This is the case, and natural radioactivity, as discovered by Becquerel in 1895, is due to this fact. It may also be expected that bombardment of elements by alpha particles as initiated by Rutherford might create again some of the combinations which have long since disappeared in the progress of the universe. This also has proved to be true. Curie and Joliot in 1933 were able to produce  $N^{13}$  by the reaction



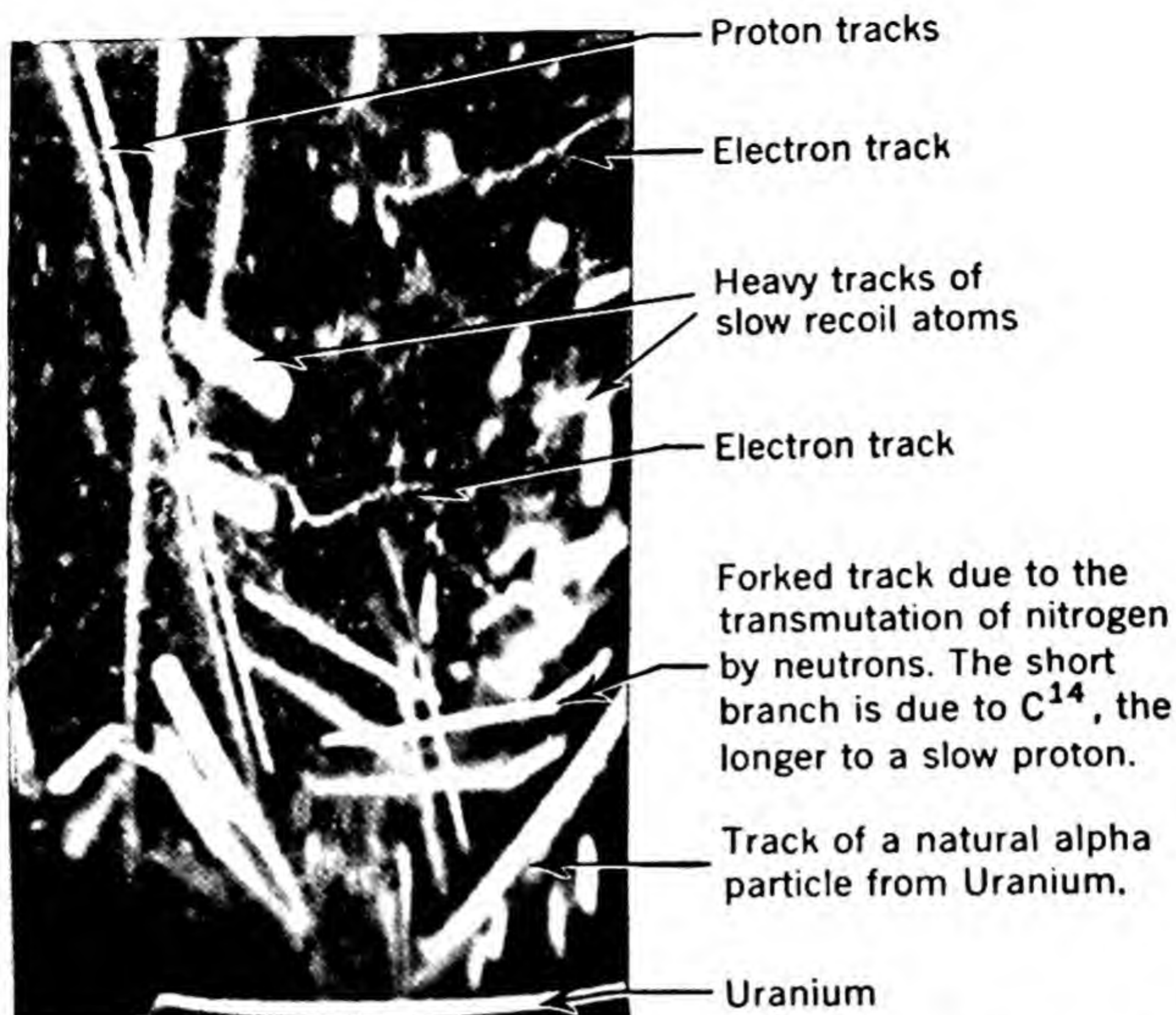
The  $N^{13}$  has too many protons and changes one of them into a neutron with emission of a positive electron. Today several hundred artificially radioactive nuclei are known.

This simple semihistorical introduction presents the three salient facts of nuclear physics: first, the formation of nuclei by combination between neutrons and protons due to strong forces; second, the existence of stable combinations of neutrons and protons due to reasons only partly understood; third, the ability of neutrons and protons to interconvert into one another with emission of electrons of some kind. We can now proceed to develop the subject.

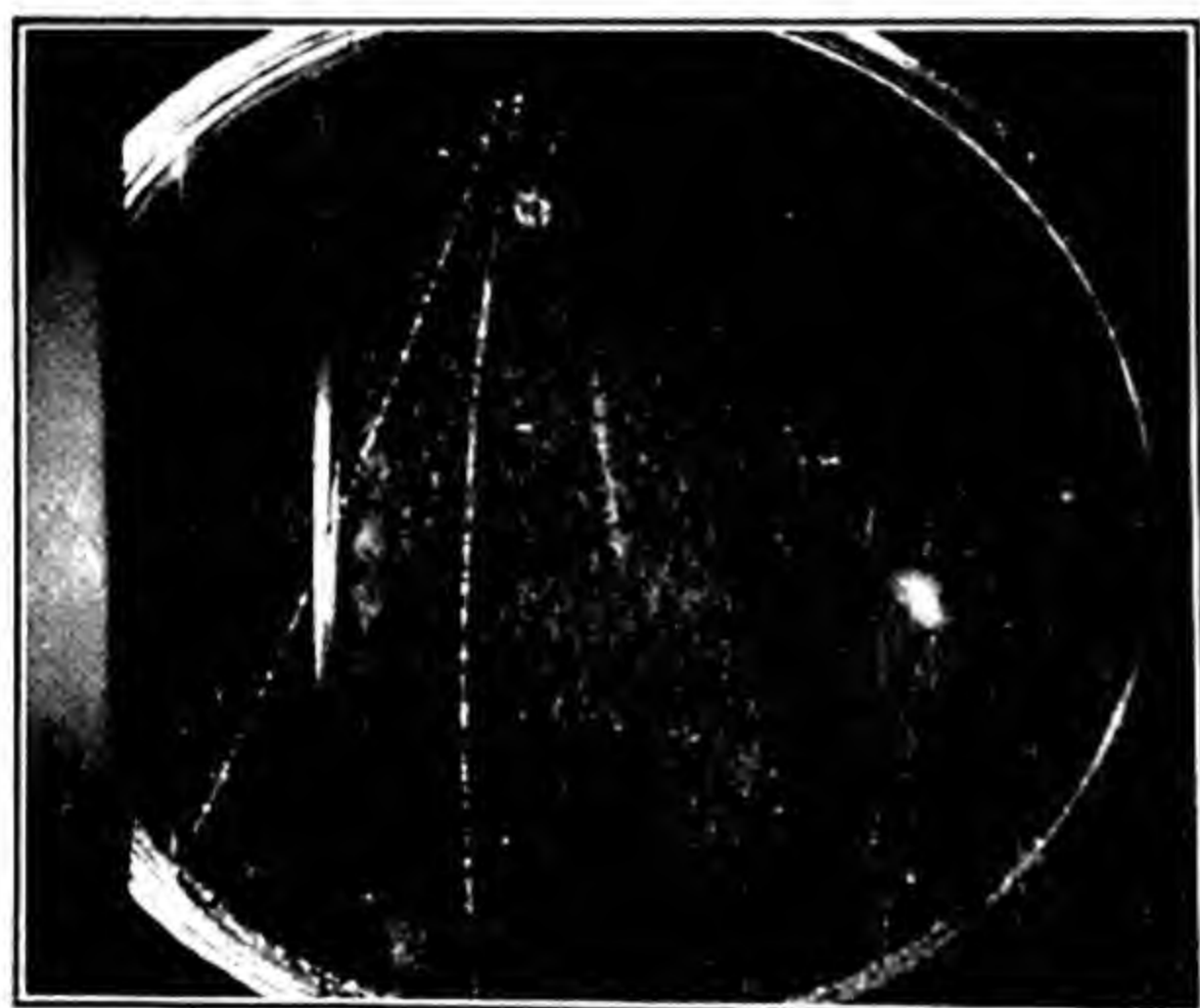
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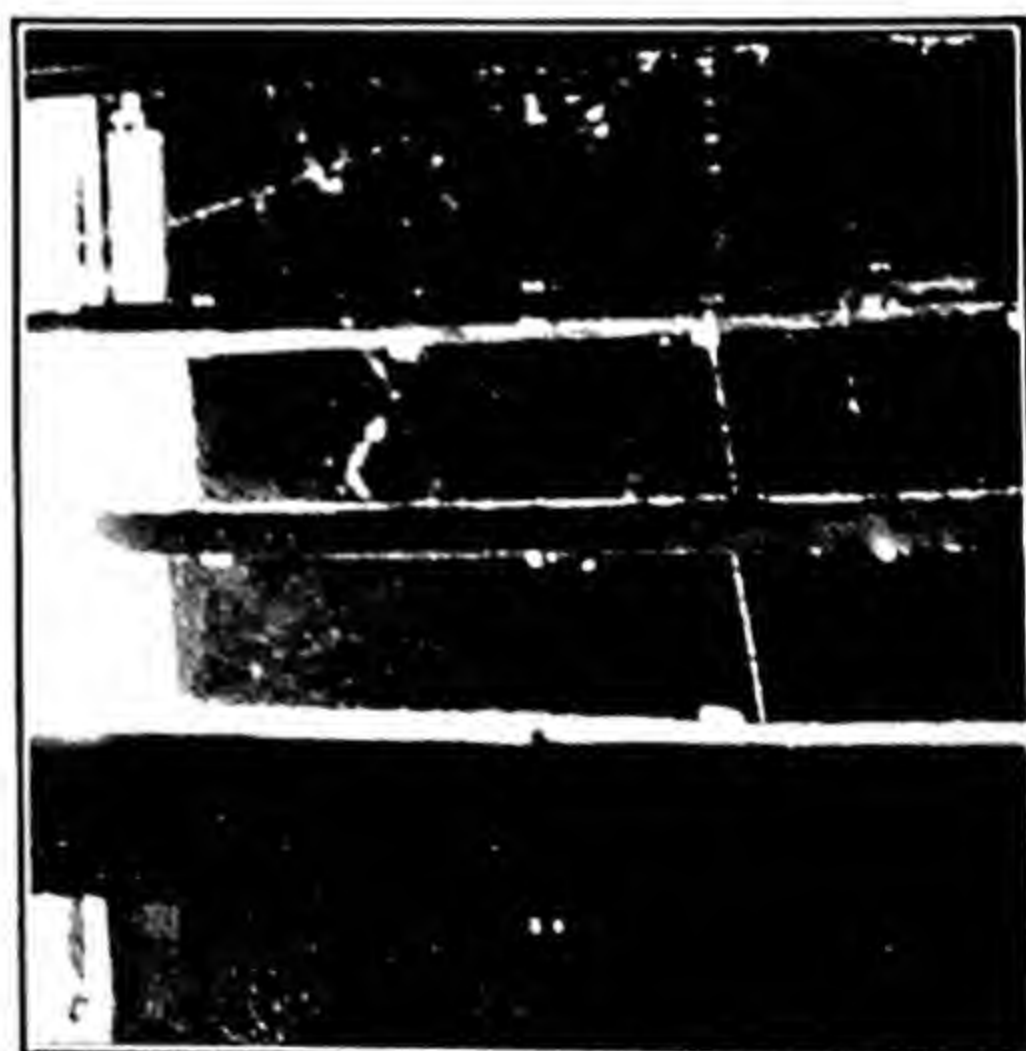




I. Reproduction of a cloud-chamber photograph, by Dr. I. A. Getting, showing the effects of a neutron source on the air and water vapor gas mixture. The track of a natural alpha particle from uranium is also visible.



II. Cloud-chamber photograph, by Dr. J. C. Street, of a pair of electrons moving in a magnetic field.



III. Photograph, by Dr. R. Sard, of a meson traveling through several layers of absorption.



## 2. Properties of Nuclear Radiations

Before proceeding to describe the results of actual transmutation experiments, and at the risk of tantalizing the reader, we shall give a short account of the properties of nuclear radiations as a help for the later chapters. We also reproduce a plate showing the tracks of electrons, positrons, protons, alpha particles, and mesons photographed in a cloud chamber. The shrewd reader will at once note that the particle itself is never pictured but only what it does. The alpha particle and proton ionize very strongly; the electron and positron very weakly; mesons produce intermediate ionization. The neutron, neutrino, and quantum produce little or no ionization directly.

Such a picturing of nuclear radiations serves the purpose of differentiating ionizing from non-ionizing radiations, but it is of no use unless coupled with a clear understanding of the nature of each radiation. We will therefore begin by subdividing the radiations into two categories, material and non-material. (Even this division is hard to make.)

The material list is headed by the *proton*, the nucleus of a hydrogen atom, having a single positive charge equal to that of the electron ( $4.80 \times 10^{-10}$  esu), a mass of  $1.660 \times 10^{-24}$  gram, or on the scale of atomic weights, with neutral oxygen as exactly 16, a weight of 1.0076. The proton, like any heavy charged particle, possesses the remarkable feature of a well-defined path in any material. This total path is spoken of as its *range*, and the range varies with the energy of the proton very rapidly but according to no simple law. The ranges of protons of energies from 1 to 20 million electron volts (Mev) \* are

\* The atomic physicist uses as an energy unit the energy acquired by an electron in falling through a one-volt potential difference, and this is called one electron volt. One electron volt is  $1.6 \times 10^{-12}$  erg. This can be compared with the chemist's method of stating atomic or molecular energies, namely, in calories per mole. If the energy of one electron volt is possessed by all the molecules in a gram-molecular weight ( $6.03 \times 10^{23}$ ), the resulting energy is 23,000 calories. Thus an electron volt per molecule is equivalent to 23,000 calories per mole.



given in Table 1 (p. 20). The definite range is a simple matter to explain in a general way: a heavy charged particle loses on the average 33 electron volts of its energy for every ion it produces and hence will produce very nearly the same number of ions for the same initial energy. Since a heavy particle is hardly deviated from its path (except by rare close collisions with nuclei) this constant amount of ionization means a constant distance traveled. The distance traversed is not simply related to the initial energy because a fast proton does not ionize so readily as a slow one.

Second on the material list is the *neutron*. This is an uncharged particle, virtually equal in mass to the proton, though actually slightly heavier, being of atomic weight 1.00899. It produces no primary ionization but may be detected by the fact that in collisions with charged nuclei it will impart energy to them and these secondary moving nuclei will ionize and be detectable. A neutron can, at best, give up all its energy to a hydrogen nucleus, giving rise to a proton whose range will measure the energy of the original neutron. Collisions with heavy nuclei cause relatively little transfer of energy from neutron to nucleus: the amount can be calculated by methods used in elementary impact problems. A useful figure to remember is that, *on the average*, a neutron retains  $1/2.7$  of its energy after a single collision with a hydrogen nucleus. Neutrons are unstable, passing into a proton and an electron after an average life of about 20 minutes.

Neutrons and protons together comprise all nuclei, and hence combinations of the two in all imaginable forms can be considered as nuclear particles. Of these, two will be selected for mention: the *deuteron* and the *alpha particle*. The *deuteron* is a proton plus a neutron held together by a strong force. Its importance lies in the fact that it can readily be accelerated by a cyclotron and that it is a potent agent in causing transmutations. The *alpha particle* is a nucleus of helium, or two protons and two neutrons held together extremely tightly. It is also readily accelerated in a cyclotron to enormous energies and in addition is of interest since natural radioactive elements emit alpha particles in amounts which just barely permitted the discovery of transmutations to be made.

The third components of the material list are electrons, positive and negative. Of these, only the negative is permanent, the positive ultimately changes, giving birth to two quanta of gamma radiation. The electron is the fundamental atom of electricity; that its charge is labeled negative is pure accident. The mass of the electron is small



compared to the proton and neutron, being  $\frac{1}{1840}$  that of the proton. This small mass renders the properties of the electron considerably different from those of the proton, first, because it is easily deflected (or "scattered") in collisions with nuclei, thus causing its path to be tortuous; second, because it is less effective in disturbing the electrons in atoms, thus reducing the ionization per centimeter; and third, because it is easily deflected in a magnetic field. The small mass of an electron also means that at high energies its velocity is very close to that of light and so, according to the theory of relativity, its mass is greater according to the relation

$$m = \frac{m_0}{\sqrt{1 - (v^2/c^2)}}$$

and its kinetic energy is

$$\frac{m_0 c^2}{\sqrt{1 - (v^2/c^2)}}$$

where  $m$  = its new mass.

$m_0$  = its "rest" mass.

$v$  = its velocity.

$c$  = the velocity of light.

An electron from  $B^{12}$ , for example, having an energy of 12 Mev has a velocity of 0.999 that of light and a mass roughly 24 times its rest mass. This fact, of changing mass, must be remembered in making calculations about electrons and their motions.

Positive electrons, or positrons, are similar to electrons in all respects but two, the sign of their charge and their longevity. Within a fraction of a second, a positron always disappears, if any material is near, and in its place two gamma quanta appear. There is a close relation between the frequency of the gamma-ray quantum and the rest mass of the positron, namely,

$$m_0 c^2 = E = h\nu$$

where  $E$  is the "energy" of the quantum,  $\nu$  its frequency, and  $h$  is Planck's constant. This interesting relation will be discussed shortly.

Of increasing significance in nuclear physics are *mesons*, or *mesotrons*. (The international conference to settle which of these words to use has not yet happened and it is largely a matter of taste. We prefer *meson* as it is shorter.) Mesons were discovered to be a component of cosmic radiation, produced in the earth's atmosphere by a collision between the primary radiation of cosmic rays and some atom.



Mesons, which are positive and negative, have gradually become more definite, and a rapid step forward was made by Gardner and Lattes in 1948 when they discovered that one form of meson is emitted by nuclei under high energy alpha particle bombardment. Mesons have a very ephemeral existence. Of those best studied, the heavier form, the  $\pi$  meson of mass 286 electron masses, decays into  $\mu$  mesons in  $10^{-8}$  second if it is given a chance. Only the positive form is usually given a chance, for the negative is attracted into nuclei with such strength that so far its main effects are nuclear explosions, known as stars. The lighter form, the  $\mu$  meson of 198 electron masses, decays into an electron in  $2 \times 10^{-6}$  second on the average. Pictures of these processes appear in the last chapter, where a fuller discussion of these most interesting particles is given.

There are possibly some neutral mesons, sometimes called *neutretos*. The difficulty of studying the nature of charged mesons is already great, and the knowledge of neutral mesons will have to wait until the technique of high energy nuclear bombardment has progressed further. Very rapid expansion of our knowledge of all kinds of mesons can be predicted in the next few years.

Mesons so far have not played a dominant role in applied nuclear physics. They are undoubtedly concerned with the nature of nuclear forces and probably also with the process of radioactivity. How, has not yet been discovered.

There are just two types of non-material nuclear radiation. The first is *gamma* radiation. This is generally classified as electromagnetic radiation of very short wavelength (and hence high frequency) so that officially one is expected to imagine gamma rays as trains of waves in which a varying electric and magnetic field is propagated through space. In actual fact this picture is considerably modified in nuclear physics. The frequency of the gamma rays is so high that their behavior is almost entirely understood by considering them as quanta of energy such that  $E = h\nu$ . This quantum of energy, like the neutron, is itself undetectable, but fortunately a gamma ray has three very strong methods of interaction with matter which make its detection relatively simple. In the first place, a collision with an electron will project the electron in the original direction of the quantum according to the ordinary rules of impact (the quantum being considered to have a momentum  $h\nu/c$ , where  $c$  is the velocity of light). In this



way the electron can acquire enough energy to ionize and be detected. Also, the maximum energy of these projected electrons is a measure of the energy of the gamma-ray quantum striking them. This phenomenon, known as the *Compton effect* (the electrons are known as Compton recoils), is the most common method of interaction between gamma rays and matter.

The second method of interaction is the *photoelectric effect*. The electric field of the gamma radiation produces a force on the electrons in an atom. While the electric field is there, a kind of ephemeral atom exists in which not all the electric field is of the usual atomic character. During this time there is therefore a chance that one of the electrons may change from a familiar bound condition (an atomic energy level) to some very different condition (external to the atom). When this occurs the quantum communicates all its energy to the electron, detaching it completely and giving it kinetic energy of motion equal to the original quantum energy less the energy of binding of the electron in the atom, which is a small correction for these energetic quanta. Again the ionization produced by the electron is the secondary effect which renders the detection of the gamma ray possible. This photoelectric effect accounts for less than one-fifth of the energy exchanges by an average gamma ray.

The third method of interaction is the strangest and most interesting. It is known as *pair production*. A short account of this is worth while. It has been noticed already that the electrons emerging from nuclei move so rapidly that their mass changes in accordance with the theory of relativity. Now if we seek to apply the theory of relativity to the motion of electrons we are bound by the requirement that, no matter what speed the coordinate system has, the laws of motion of the electron remain unchanged. This is a simple requirement, but, when mathematical expressions including both the motion of the electron and the electric field are formulated, it is found that the equations contain two solutions for the kinetic energy of the electron: one positive; the *other negative*. A moment's thought will show that negative kinetic energy does not mean anything that we actually observe, since the quantity  $\frac{1}{2}mv^2$  must automatically be positive, and this presented Dirac, the formulator of this theory, with a first-class dilemma. With unusual courage, Dirac retained his theory by proposing that these states of negative kinetic energy, though not observable, still exist; they exist but are normally completely filled with electrons. Since all the possible conditions in which these negative



kinetic energy electrons can find themselves are already satisfied by electrons, we can never detect any change in them and so do not notice their presence. This line of reasoning, however, does not affect a gamma ray, to which these electrons are no more than electrons and therefore capable of being influenced. By a process similar to the photoelectric effect the gamma ray may give energy to these electrons; and, if it does, an electron in a state of negative kinetic energy may acquire enough positive kinetic energy to detach it from its state of negative kinetic energy and release it. Thus a *negative electron would appear*. At the same time there would be a hole in the completely filled states of negative kinetic energy, and this hole, or absence of an electron, will appear like a positive electron except that it will vanish when the hole is filled.

The theory as simply outlined above lacks one important feature: we must include in the kinetic energy (whether positive or negative)

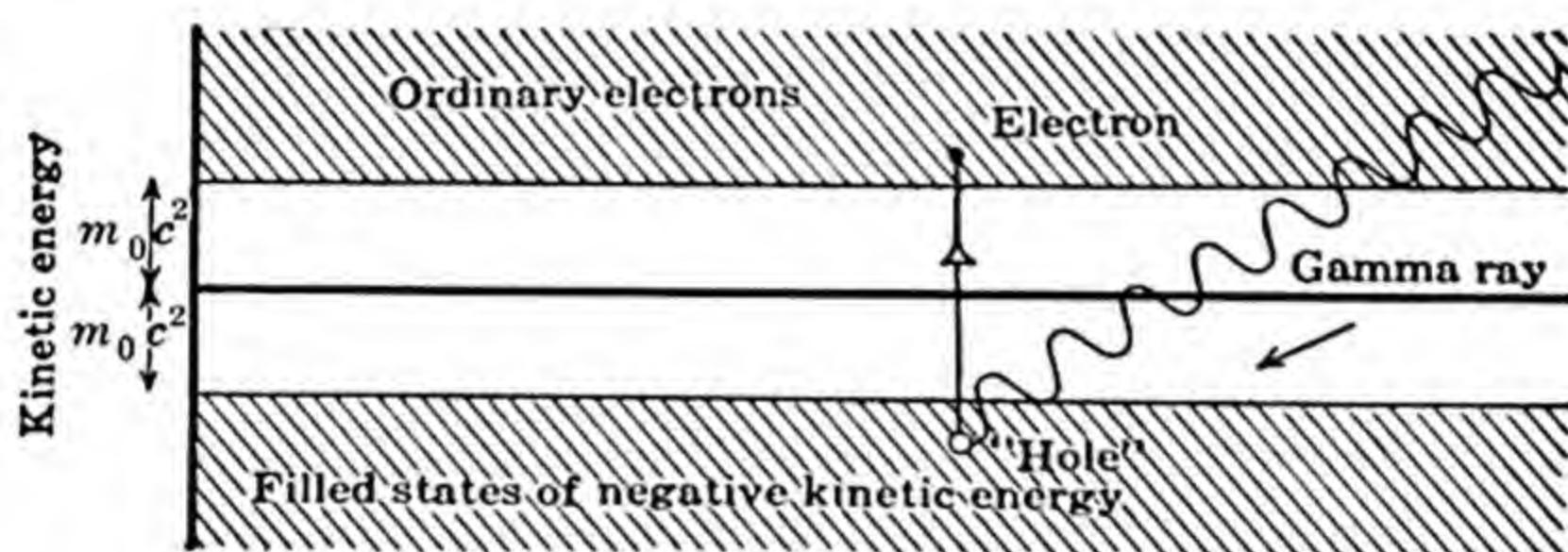


FIG. 1. Indication of the process of pair formation. The electromagnetic field of the gamma ray supplies enough energy to an electron in a state of negative kinetic energy to eject it, leaving a "hole" which behaves like an electron of positive charge.

the term  $m_0c^2$  which corresponds to energy possessed by a particle in virtue of its mass alone. We can now picture all the electrons we wish to consider as in two bands, as drawn in Fig. 1. The process of pair formation is indicated schematically; notice that a pair cannot be formed unless the quantum has an energy greater than  $2m_0c^2$ , which is almost exactly 1 Mev.

The discovery of the positron and of the process of pair formation was one of the most startling confirmations of one of the strangest theories in science. Pairs are found only when quanta having an energy in excess of 1 Mev fall on matter; the chance of their formation increases with the quantum energy and also with the charge of the nucleus of the material from which they are derived. Thus for 3-Mev



gamma rays impinging on lead, nearly one-half the energy lost by the gamma quanta is in the form of pair production. For aluminum the amount is much less, being only a few per cent.

These three processes contribute to the absorption of gamma rays in matter. To screen out gamma rays one therefore needs an inch or two of a heavy element such as lead. This is in contrast to screening neutrons which requires careful design involving walls at least 3 feet thick, 10 feet being much safer.

While the subject of pair production is still fresh, the reverse process of *annihilation* should be mentioned. This is the ultimate fate of every positron. The hole in the negative energy states becomes filled up by an electron from the positive energy states. This means that an amount of mass equal to two electron masses disappears, and since, if mass is annihilated some form of energy must appear, gamma radiation is evolved. This process must, however, also permit momentum to be conserved, and as there is no momentum to begin with, there must be no momentum at the end, a condition which is realized by the ingenious process of producing *two* gamma rays of equal energy, moving in opposite directions. Thus when a positron is annihilated, two quanta, of energy each equivalent to  $m_0c^2$ , are evolved. Thus any radioactive element which emits positrons also emits a secondary gamma ray, the *annihilation radiation*, whose energy is almost exactly 0.5 Mev. This phenomenon has also been observed, adding further confirmation to the theory for which Dirac received the Nobel prize.

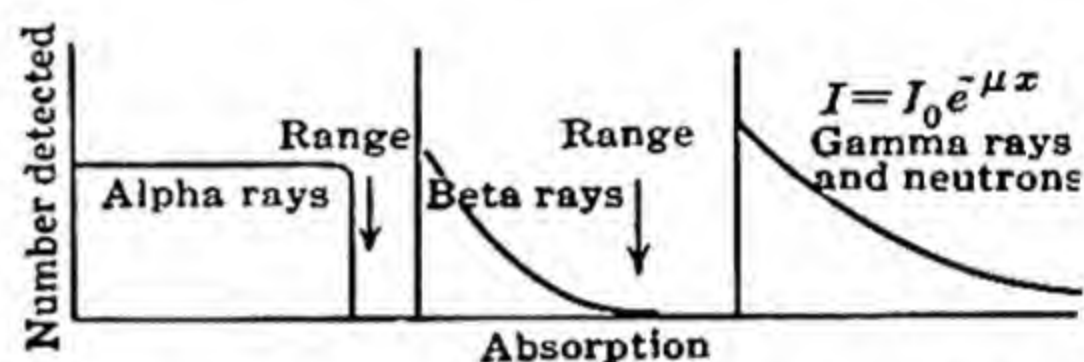


FIG. 2. Absorption curves for alpha, beta, and gamma rays and neutrons. Alpha and beta rays have a definite range; gamma rays and neutrons are absorbed exponentially.

The second non-material radiation is the *neutrino*. The neutrino is a particle which is supposed to share in the energy balance of a radioactive change; its rest mass is supposed to be zero, it is neutral, and its only possible detectable influence would be an ion pair every mile or so of its path. This is too small to observe. It occupies much the same sort of place in physics that the ether did in the nineteenth century—much talked about but never observed. A new theory may dispel the actual existence of the neutrino, which is hardly true of any of the other radiations yet discussed.



Before concluding this chapter a few odds and ends should be collected. The three terms alpha, beta, and gamma rays are applied, respectively, to helium nuclei, to electrons emerging from nuclei in radioactive transformations, and to quanta. They are in decreasing order of mass and in decreasing order of absorbability. Alpha and beta rays have definite *ranges*; they may be stopped completely by relatively small amounts of materials. Gamma rays, like neutrons, can only be diminished in numbers by absorbers, never wholly cut out. The three types of absorption curve are shown in Fig. 2. The beta rays emitted from a radioactive substance are not homogeneous in energy, yet a definite range is always observed. In Table 1 are shown the ranges for protons, deuterons, and alpha and beta rays of various energies. The thicknesses of materials actually traversed by these rays are so small that they cannot easily be measured directly. It is very simple to express the thickness as the *mass per unit area* of a given absorber, for then accurate weighing permits an accurate figure to be given. This procedure is universal in nuclear measurements, so much so that the authors find it easier to visualize a thickness stated in milligrams per square centimeter than directly in fractions of a millimeter. It has one other advantage; it is more directly related to the fundamental absorbing process—the number of electrons encountered—than the thickness, which may vary considerably from element to element for the same range ray. This variation in actual thickness is due to the varying density of the materials, a factor which does not appear if the absorption is estimated by weighing. All the figures here given apply to aluminum as absorber.

TABLE 1

ALL RANGES IN MILLIGRAMS PER SQUARE CENTIMETER OF ALUMINUM

Energy (Mev)	0.5	1	2	3	4	5	7	9	10	15	20	30
Proton range	1.3	3.5	10.6	19.6	35.0	50.1	90.1	143.0	174.0			
Deuteron range	1.1	2.6	6.9	13.3	21.2	32.4	55.9	86.0	100.0	205.0	348.0	720.0
Alpha-particle range	0.5	0.8	1.6	2.1	3.8	5.3	8.9	13.5	15.6	39.2	50.1	102.0
Beta-ray range	111	383	926	1470	2010	2650						

In this chapter we have described the products that can be emitted from nuclei, dividing them into two categories, material and non-material. The material includes protons, neutrons, positive and negative electrons, and positive and negative mesons; the non-material,

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quanta and neutrinos. The great game of composing matter must be played with these; whether any combination will work, once formed, depends on the laws of force which operate between the parts of the combination, and so on the wisdom of the choice of the constituents.



### 3. The Detection of Nuclear Particles

*—the intense atom glows  
A moment, then is quenched in a most cold repose.*

*Without exception, charge and speed are required for the detection of nuclear particles. Radiation or neutrons which possess no charge must be detected by secondary effects in which they cause charged, rapidly moving particles to appear. It is thus necessary to concentrate on the effects produced by fast charged particles in moving through matter in order to understand the means by which we detect nuclear products.*

#### ✓ Ionization

To our ordinary senses, matter, even a rarefied gas, appears continuous. This continuity is one of the most troublesome optical illusions there is. Had nature ordered things so that our eyes would focus x-rays it is likely that all modern physics would have been classical physics early in the nineteenth century and the mysteries of the atom would seem today as trite as Boyle's law. As it is, we have had to wait until experimental technique has enabled us to see how we must exert our imagination to appreciate the true nature of matter. *Matter is mostly emptiness.* Even in a solid the centers of the atoms are separated by distances a million times greater than the atoms themselves. The seeming impenetrability of a solid is due solely to the extremely powerful forces exerted by the charged atoms in the solid as soon as other charged atoms (in some foreign piece of matter such as our fist) are brought sufficiently near. If for an instant electrical charge were abolished in a championship bout the contestants would readily pass right through each other in ghostly fashion. This being so, a small, rapidly moving charge will be able to penetrate matter and, roughly speaking, it will find interference only in the electrical attractions or repulsions which it experiences.

In Fig. 1 a picture representing the passage of an electron through a gas is drawn. The electron enters at the lower left and passes near atom A. Although this atom is, in toto, electrically neutral, the outer



portion, consisting of planetary electrons, is negatively charged, and can shift with respect to the nucleus. When this shift occurs an attractive force is exerted on the moving electron. The moving electron is deflected while atom *A* recoils as indicated. The same is true in a varying degree for other atoms such as *B* and *C*, and in each such encounter the moving electron imparts momentum to atoms in the gas, momentum which is lost from its own motion. In this way the moving electron could be brought to rest and would leave in its track a host of moving atoms.

Unfortunately such moving atoms, produced in so-called elastic collisions, are not readily detectable; if this process alone were taking place our knowledge of nuclear physics would be very slight. It may happen, however, that a very close encounter with an atom takes place, in which event there is an extremely powerful repulsion between the moving electron and the atomic or bound electron, which may receive sufficient momentum to dislodge it from the atom, a process known as ionization. Such a procedure changes the condition of the atom encountered, requires temporarily the absorption of energy, and is known as an *inelastic collision*. These inelastic collisions which result in ionization afford the means by which the detection of nuclear charged particles is effected. It is found experimentally that a swiftly moving electron produces approximately one ionized atom for each 33 electron volts it possesses, which means that an electron of 1,000,000 electron volts energy will, before it is stopped completely, produce 30,000 ionized atoms. Since the ejected electron automatically leaves a positive residue, each ionization process gives two ions, or an ion pair, and hence an average electron moving with an energy characteristic of nuclear processes can produce 30,000 ion pairs.

These 60,000 charges are something we can hope to detect. It remains to consider the most expedient methods.

### Direct electrometer detection

Before taking a look at the sensitive instruments with which the physicist is equipped to measure small electrical quantities let us ex-

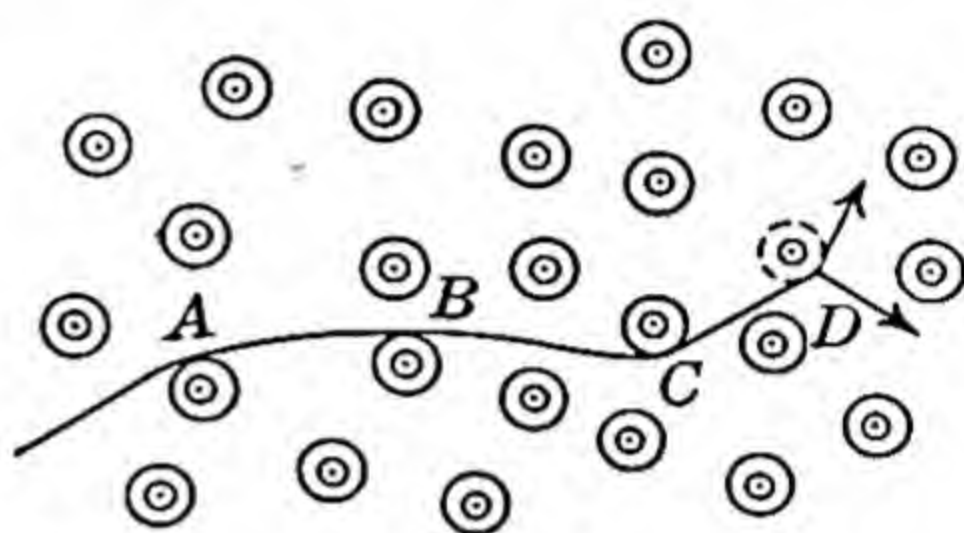


FIG. 1. Path of an electron showing repulsion when near electrons in atoms and indicating the recoiling atoms. At *A*, *B*, and *C* the collision is elastic; at *D* an electron is detached from its atom, and an inelastic collision, here resulting in ionization, takes place.



press 60,000 ion pairs in electrical units. Each ion has a single electronic charge of  $4.8 \times 10^{-10}$  esu or  $1.6 \times 10^{-19}$  coulomb, so that the total is  $2.8 \times 10^{-5}$  esu or  $9.6 \times 10^{-15}$  coulomb. One glance at these figures tells the physicist that he requires instruments of very great sensitivity. Let us briefly survey the available instruments. The first is the galvanometer; it can be designed to have a current sensitivity of  $10^{-12}$  ampere or a quantity sensitivity of  $10^{-10}$  coulomb. Un-

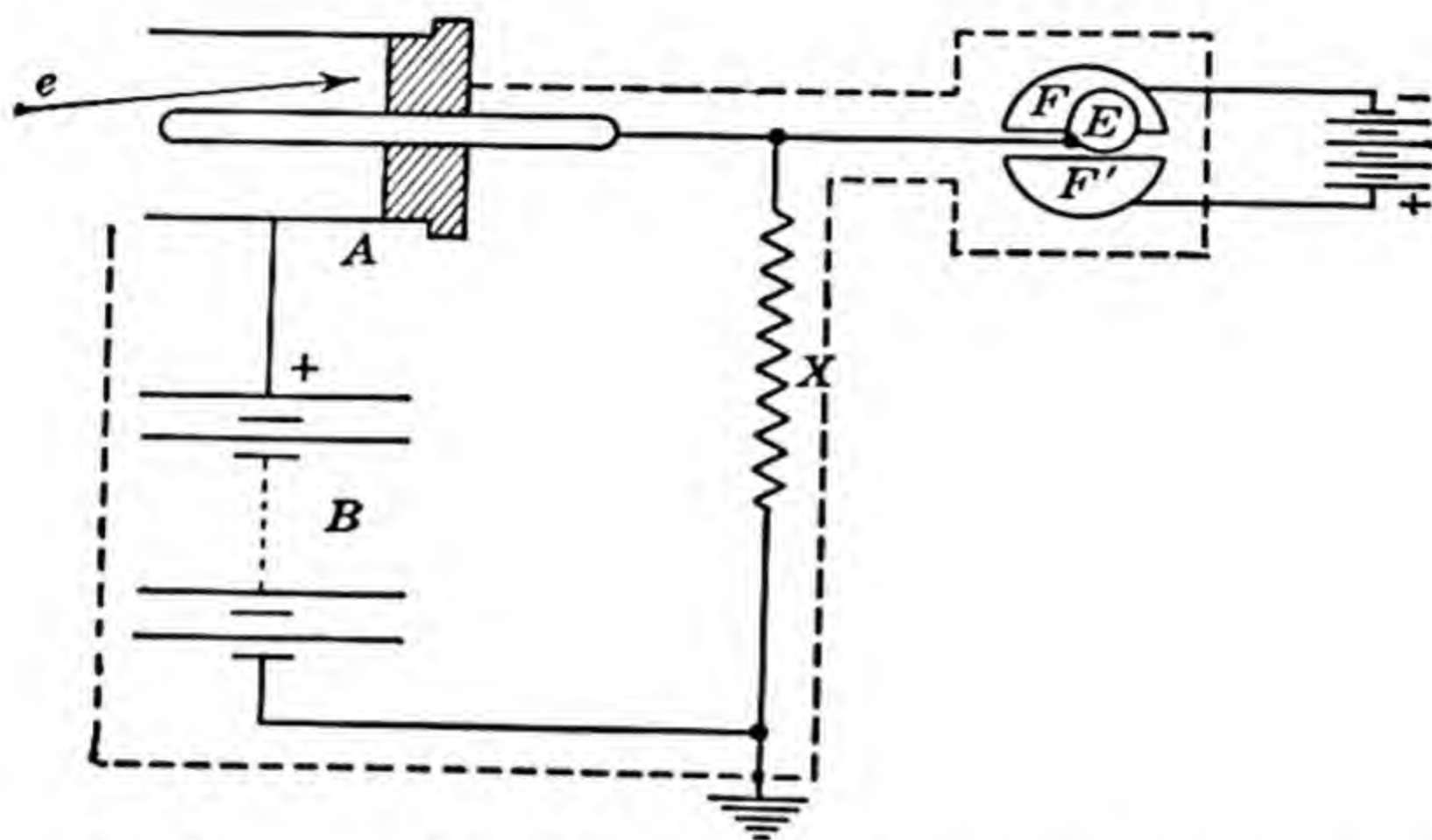


FIG. 2. Schematic diagram of ionization chamber and electrometer used for the detection of single fast nuclear particles. *A* is the ionization chamber with an insulated central electrode. Battery *B* causes the positive ions produced by the incoming fast nuclear particle *e* to be driven to the electrode and raise the potential of the movable vane *E* which is suspended by a fiber above the plates *F* and *F'*. This rise in potential causes the vane *E* to move.

aided, therefore, it is quite useless. The second is the electrometer. This can be made to have a sensitivity of  $10^{-5}$  volt per division with a capacity of  $10^{-11}$  farad. The charge it could detect, then, is  $10^{-16}$  coulomb, and it is thus possible to detect the charge produced by an electron in ionizing a gas *directly* by means of a sensitive electrometer. In practice this method, though by far the simplest in principle, is not used for reasons to be given later in the chapter, but as it shows most simply the general arrangement of nuclear detecting apparatus a diagram is given of an electrometer applied to detect single electrons.

The circuit is shown schematically in Fig. 2. The electron enters the *ionization chamber A* where it causes ions to form. The ionization chamber contains two electrodes (in this instance the outer case and an insulated central rod) between which an electric field is applied by the battery *B*, the circuit being completed by a high resistance *X*. The ions formed by the fast electron are separated by



the electric field, and the plus ions are driven to the central electrode whose potential rises when it receives the charge. The central electrode is connected to a movable vane  $E$  supported by a thin conducting fiber. This vane is suspended above two plates  $F$  and  $F'$ , which are maintained at positive and negative potentials by a battery, and normally the vane takes up an equilibrium position between  $F$  and  $F'$ , depending on its own potential, the potentials of  $F$  and  $F'$ , and the twist in the fiber. As soon as the potential of  $E$  is changed the equilibrium is disturbed and the vane swings around, its motion being observed by a mirror attached to it and a reflected beam of light. The dotted lines indicate metal casing which is used to shield all leads connected to the electrometer as the pulses caused by starting currents and so on around the laboratory can easily induce a potential of 0.01 volt, which is a hundred times larger than the effect to be observed. Screening by metal casing diminishes this "pick-up." This instrument is the "duant" electrometer, designed by Hoffmann and used in several important nuclear investigations. Attention should be given to the  $A-B-X$  section of the apparatus. The combination of a field driving a charge on to an electrode which is insulated except for a very high resistance is, in one or another variation, extremely common practice.

The reason for the infrequent choice of this method of detection lies in the electrometer, which is far from rugged and requires skill and experience to set up and keep operating. Since the fiber has to be exceedingly thin, the vane must move very slowly; this means that the period of the electrometer is long so that several seconds must elapse between the detections of charged particles. If our available source of fast electrons is strong so that we are never required to detect less than about a thousand incoming electrons per second, then the electroscope, which is much simpler and more rugged, can be used.

Almost everyone is familiar with the ordinary gold-leaf electroscope. Not everyone is aware that it has played a major role in the study of radioactivity. If a gold-leaf electroscope is charged, so that the leaves diverge, it will remain in that condition almost indefinitely. But if the space near the leaves is ionized the ions in the air are attracted to the charged leaves and in this way tend to neutralize their charge and cause the leaves to collapse. The leaves may be observed through a microscope of low magnification with a scale in the eyepiece and the rate of collapse of the leaves measured in terms of divisions on the scale per minute; this rate is a measure of the ionization current. The ionization current, in turn, is a measure of the number of incoming



electrons. This extremely simple device has probably turned out as much nuclear research data as any other instrument. It can be made reasonably sensitive by having very thin gold leaves, or more generally by using a single leaf which is repelled away from a fixed support. It does not have a linear scale, but if the timing is always carried out over the same positions of the gold leaf then it is certain that the same amount of charge has been neutralized and the differing times indicate the relative currents. If necessary an electroscope can be calibrated so as to read a current in amperes, but this is seldom necessary because it is nearly always better to compare one radioactive source with a standard source.

### The Lauritsen electroscope

The advantages of the simplicity of the gold-leaf electroscope are retained and the advantage of greater sensitivity is added in a beautifully simple quartz-fiber electroscope designed by Lauritsen. In the gold-leaf electroscope the force opposing deflection of the leaves is gravitation; in the Lauritsen electroscope the moving fiber is so thin and light that gravitation exerts virtually no effect on it. In place of gravitation as the restoring force the elastic force in the quartz is utilized; this is advantageous because it is possible to find an arrangement which will give a much more nearly linear scale for the instrument. The electroscope is illustrated schematically in Fig. 3. The heart of the instrument is shown in the small diagram (a). It consists of a small metal frame shaped as shown, with a quartz fiber (which has gold sputtered on it to render it conducting) attached to it as indicated. The end of this fiber would be hard to see, so a little T is made on the end by fastening a short piece of quartz fiber, indicated in the diagram by the dot. The T is in a plane perpendicular to the paper. If this whole system is insulated by amber and a charge given to it the repulsive force between like charges will cause the fiber to bend away from the metal support. The presence of ions in the neighborhood will cause the neutralization of the charge and a return of the fiber to its original position. As the length of the fiber is only about 6 mm, an extremely small charge is sufficient to cause considerable deflection. Even so, this would be hard to see, so a microscope with a magnification of about 50 is used to view the fiber as illustrated in the main diagram. To charge the electroscope a potential is applied to the frame by pushing the spring contact S which is attached to a source of about 100 volts, either from batteries or from a vacuum-tube rectifier. Both the leads to S should include resist-



ances of about 100,000 ohms to prevent any heavy currents from flowing if the fiber is out of place for any reason and is shorting to ground. The image of the T is made to coincide with the scale at *D*, and the passage of this image over the scale is recorded. In spite of the delicacy of the fiber, the instrument is remarkably rugged. This is because the fiber has almost no inertia and so experiences little force unless disturbed by air currents. The case effectively prevents these from being present, and the whole instrument can be carried

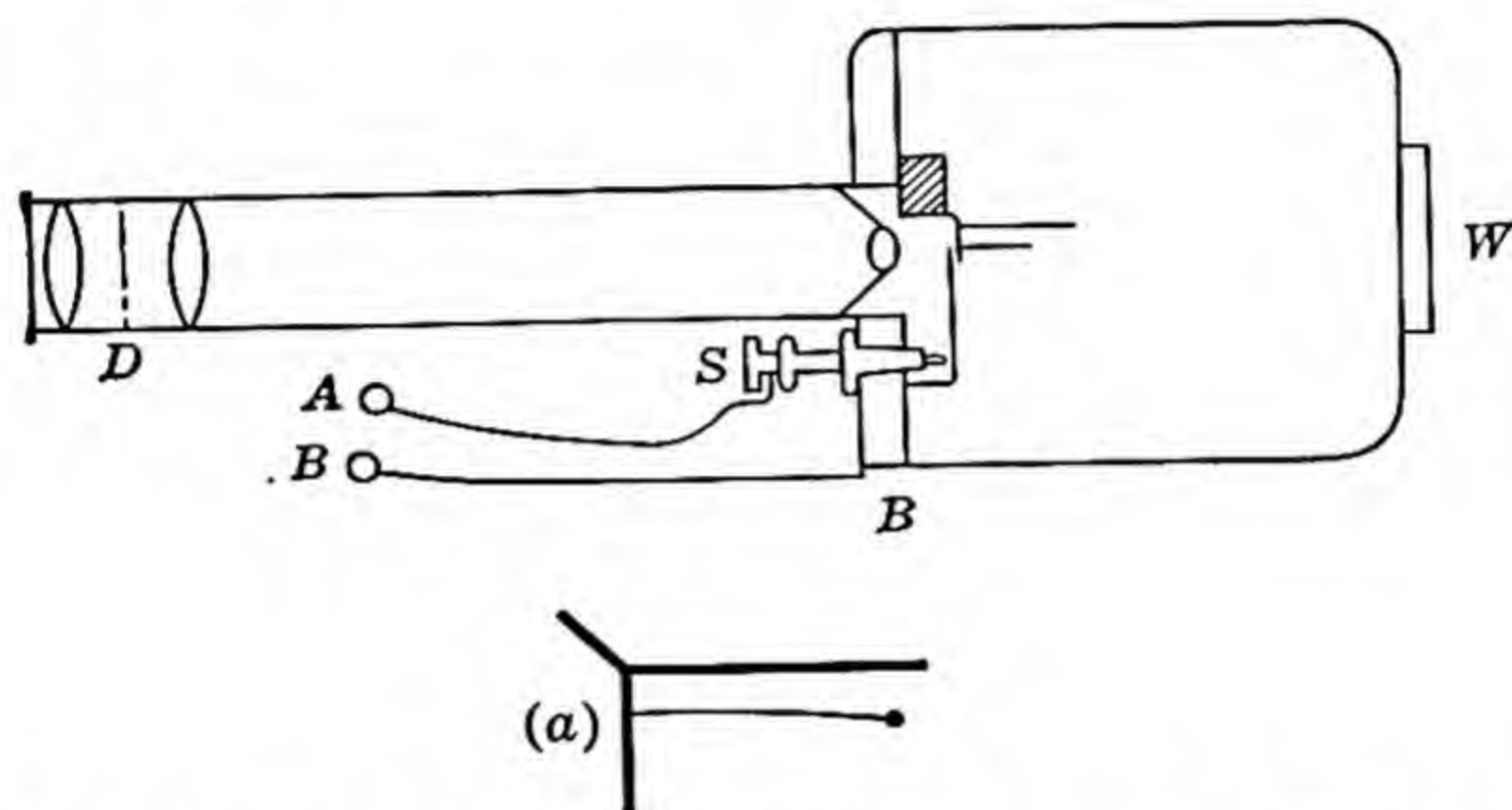


FIG. 3. The Lauritsen electroscope. (a) The detail of the metal frame and quartz fiber which is repelled from the frame when both are charged. Ionization in the space near this frame causes discharge of the frame and fiber and so a return to normal at a rate which measures the ionization current. The fiber is observed by a microscope, the illumination being through the ground-glass window *W*.

around with no more than sensible care. If it is necessary to adjust the fiber when the instrument is installed, the adjustments should be made with a handkerchief over the nose to prevent air currents. For the same reason the electroscope should be installed in some location where the temperature is reasonably uniform as convection currents may make the T ride off the scale. The most likely sources of temperature irregularities are the light used to illuminate the fiber, and the vacuum-tube rectifier. These must be kept at a little distance from the instrument. The scale is nearly but not quite linear; it is still necessary to carry out the timing over the same scale divisions at each observation. The sensitivity is such that a millicurie at a distance of a meter produces a motion of about five scale divisions per minute, and the background in the absence of radiation is about the same number of divisions per hour. If time is available it is possible to detect as few as ten electrons per minute.



### Application of vacuum tubes: the "linear amplifier"

If we return to our survey of physical apparatus available to detect the ionization produced by a swiftly moving particle we find that we have to consider the vacuum tube. It proves to be a potent aid. The vacuum tube has the tremendous advantage of utilizing atomic particles themselves to detect the ionization due to other atomic particles. A stream of electrons is virtually without inertia and hence extremely susceptible to any electrical influence. It is fundamentally this fact which has made the vacuum tube practically indispensable in detecting small amounts of radiation.

Consider the circuit of Fig. 4. Here the ionization chamber, battery, and high resistance are the same as before, but in place of the

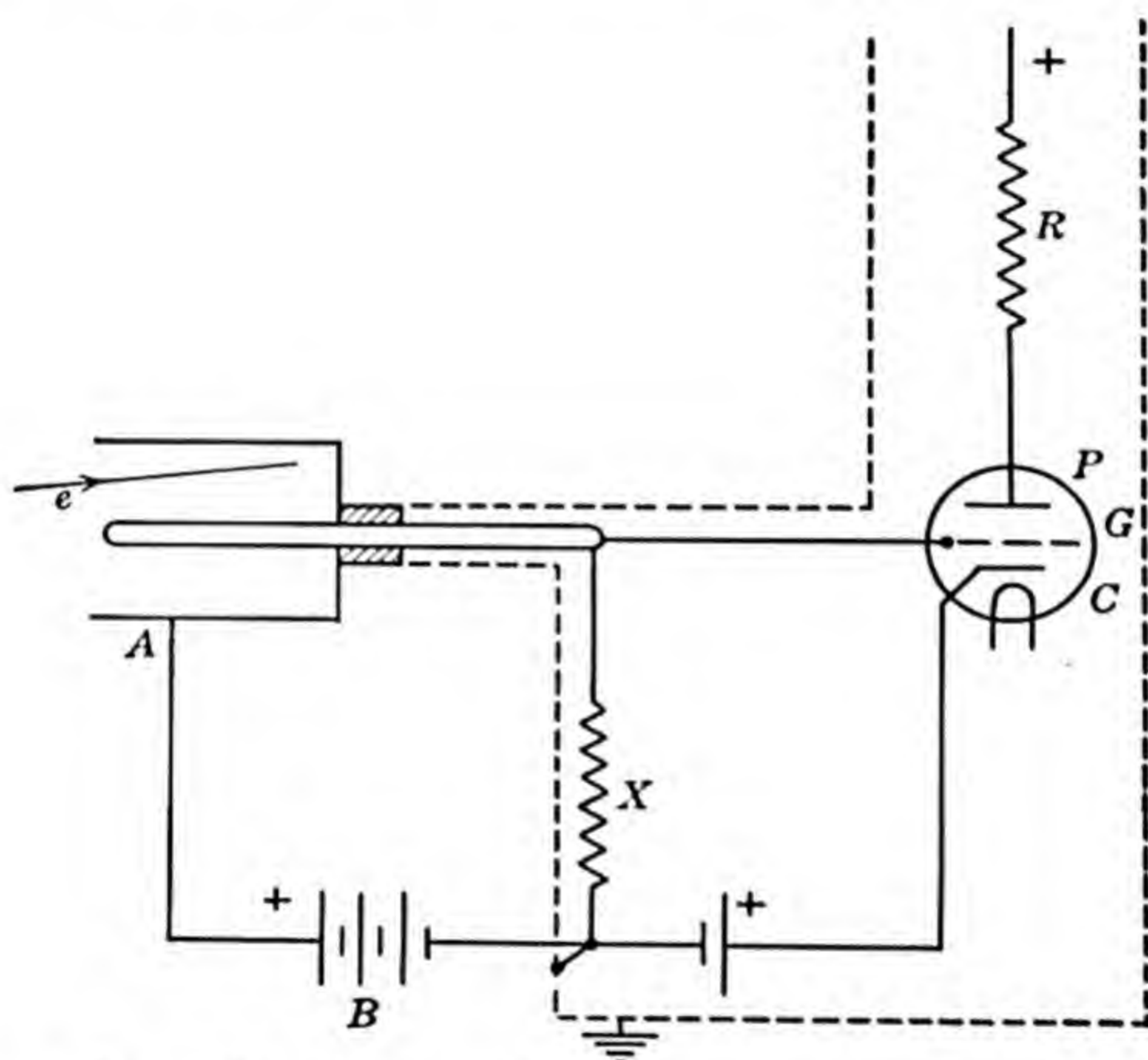


FIG. 4. Vacuum-tube and ionization-chamber arrangement. The rise in potential of the central electrode is communicated to the grid  $G$  of the triode, causing a change in the plate current and hence a relatively large potential difference across the plate resistance  $R$ .

electrometer the grid of a triode is connected to the central electrode. As before, the collection of charge on the central electrode causes a rise in its potential and hence a rise of the potential of the grid of the vacuum tube. This causes an increased flow of electrons *produced in the cathode  $C$*  to the plate  $P$  and from there through a resistance to the plate battery. The ionization chamber thus effects control of a separate electron stream, and with a well-designed triode the potential



difference produced across  $R$  by the momentary surge of electrons to the plate can be twenty or thirty times as great as the initial rise of potential of the grid. This potential difference could be used to operate a relatively insensitive electrometer, but in practice it is far better to apply it to a second vacuum tube, and so on, until the amplification of the initial pulse is a hundred thousandfold. The voltage so produced at the output is then 10 volts or so and may be detected in almost any way we please, for example by listening to the "cracks" in a loud speaker or observing the deflections of a cathode-ray oscilloscope. This is the method of the "linear amplifier," widely used in research in pure nuclear physics. Such an amplifier is not easy to operate, though far more rugged than an electrometer, and even though it affords the best method of detecting the primary ionization of a proton or alpha particle it is still not the best method for ordinary use. In fact, careful consideration shows that it is best to *change the events in the ionization chamber* to secure the simplest means of detecting the ionization due to a single particle.

### The proportional counter

Returning to our picture of an electron moving past a series of atoms and occasionally "knocking off" other electrons in its flight, thus causing ionization, let us consider the fate of these freshly detached electrons. In the absence of a strong driving force—an electric field—they will drift around until they encounter a positive ion with which they recombine to form a neutral stable atom, and that concludes the adventure. This may be drastically altered if a strong electric field is imposed on the electrons, for then they will immediately be accelerated and in a strong enough field may acquire so great a speed before an encounter with a neutral atom that they can in their turn produce ionization. Notice that the farther they go before meeting an atom the better is this chance, for they gain speed continually until checked by meeting another atom. Now this ability to ionize will also be possessed by the electron detached in the ionization process, and so on, a very rapid accumulation of electrons taking place, which can reach a thousandfold greater number than the initial ionization. This process of *ionization by collision* is exactly what we have been seeking, for by using it skillfully we can increase the initial charge to be detected by a factor of as much as a thousand, thus reducing the amount of amplification required from a vacuum-tube amplifier to the point where a simple, stable, easily constructed amplifier will suffice for the detection of the incoming electron.



A circuit is shown in Fig. 5 which will detect single fast electrons very readily. The very strong field is produced in the neighborhood of a fine wire. The initial electrons are pushed toward the wire by the negative field, and, if they arrive there before they have recombined (as they will in a high field), they produce ions by collision near the wire and consequently a multiplication of charge collected by the wire.

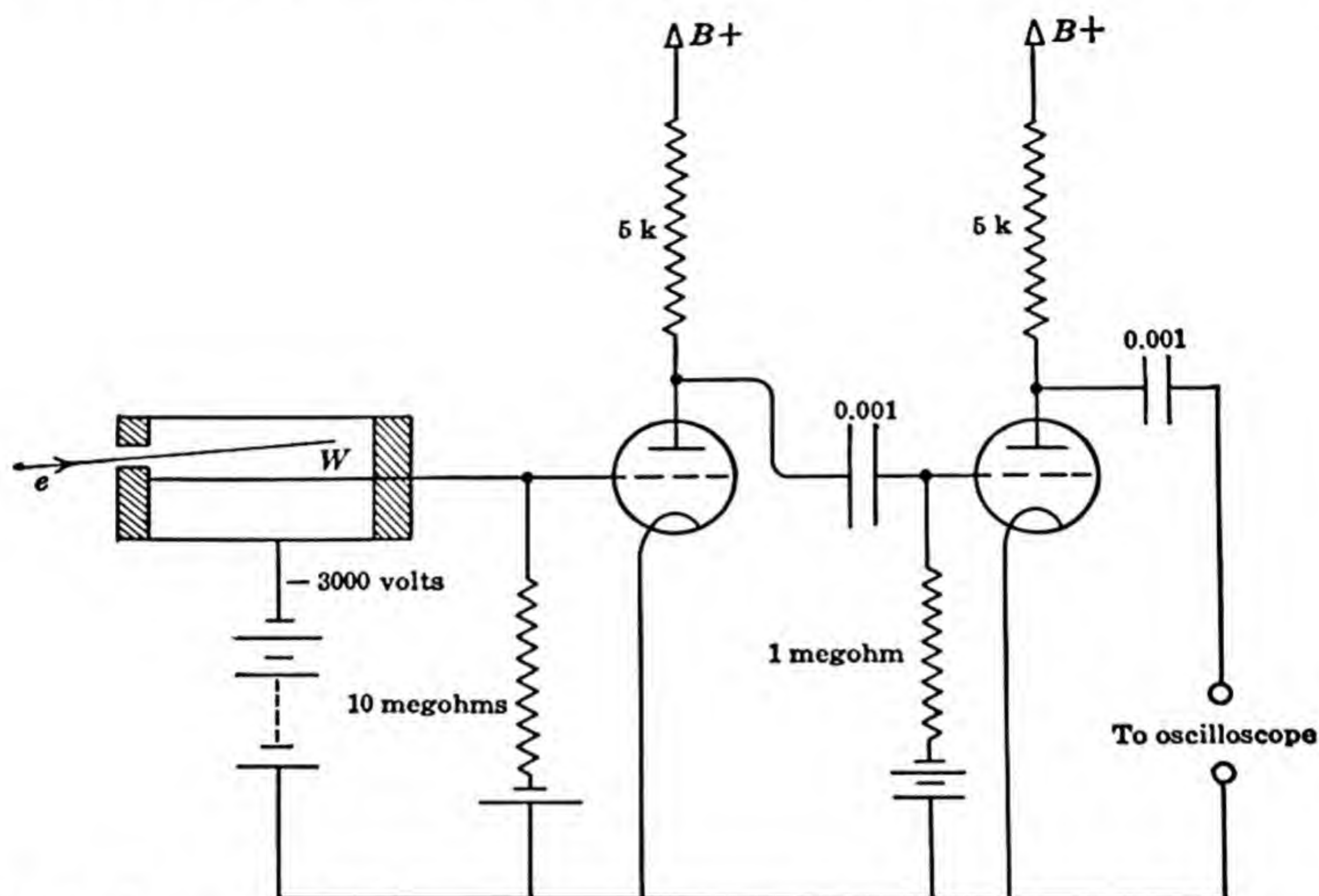


FIG. 5. Proportional counter with amplifier. A fine wire is stretched axially in a cylinder to which is applied a high negative voltage. The electric field near the wire is so high that it causes ionization by collision, which multiplies the charge collected by the wire manifold and renders a two-stage amplifier sufficient to operate an oscilloscope.

This multiplication of charge is called "gas amplification." A simple two-stage amplifier is sufficient to operate an oscilloscope. If the space around the wire is partially evacuated, the distance between atoms is increased and the process of ionization by collision can be made to occur for a lower applied voltage.\*

This is frequently done. Such a device is known as a *proportional counter*, so called because the charge collected by *W* is roughly proportional to the initial ionization. It is excellent for detecting alpha

\*The diagrams are for purposes of explanation only. For actual construction the references at the end of the chapter should be consulted.



particles and protons. A modification, due originally to Rutherford and Geiger and known as the *Geiger counter*, or, sometimes, in honor of a later developer, the Geiger-Müller counter, however, supersedes it for detecting electrons. Historically the Geiger counter was devised before the proportional counter, but as it is rather more complex it is better to consider it later. The Geiger counter calls on more than the process of ionization by collision; it utilizes some of the phenomena of the discharge. The proportional counter will count at higher rates than a Geiger counter.

### The Geiger-Müller counter

If it were true that no available electrons resided in the metal wire or metal case, then no matter how high the field applied (within some limits) there would never be more than a momentary rush of ionization following the arrival of the electron. This is not true, however. All conductors contain electrons, some of which can be extracted in various ways. If these electrons are, for some reason, pulled into the gas, then they will ionize by collision and a continuous discharge will result, maintained by fresh electrons from the metal. There are various reasons for the attraction of electrons from metal to gas; one is the fact that, in the process of recombination which is always taking place, light is emitted. In the language of modern physics we refer to this light as a number of *photons*. These photons are able to cause the emission of photoelectrons from the surface of the counter wall, and these photoelectrons can then keep the discharge going. A second reason is the hitherto neglected positive ion, which is simply the rest of the atom after the electron has been ejected. These ions are heavy, move slowly, and hardly ionize at all; but when they approach a metal, at the moment before they make contact, they exert a great outward force on electrons in the metal and may pull out several before being themselves absorbed by the metal. The nature of the surface has an effect on the number of electrons pulled out, no matter how the actual ejection is produced. If a sufficiently high field is applied between the electrodes in the counter the emission of these electrons from the walls will cause a discharge which will continue permanently so long as the voltage across the tube is kept there.

It is thus easy to see that, if a single electron arrives in a gas across which a very high field is maintained, a discharge may result in which a very large flow of current may take place. The electron thus acts as a trigger which sets off the latent discharge. Obviously such a method of detection is exceedingly sensitive. The reader will at once,



however, inquire why the discharge should cease, and the consideration of this matter is of great importance in understanding the operation of the Geiger counter.

If we suppose that the two electrodes are connected to a large source of power the discharge may well be maintained for an uncomfortably long time and would be quite dangerous. On the other hand, if we imagine that the two electrodes are first charged and then insulated before the "trigger" electron appears, the discharge will cease when the difference of potential between the two electrodes falls below that

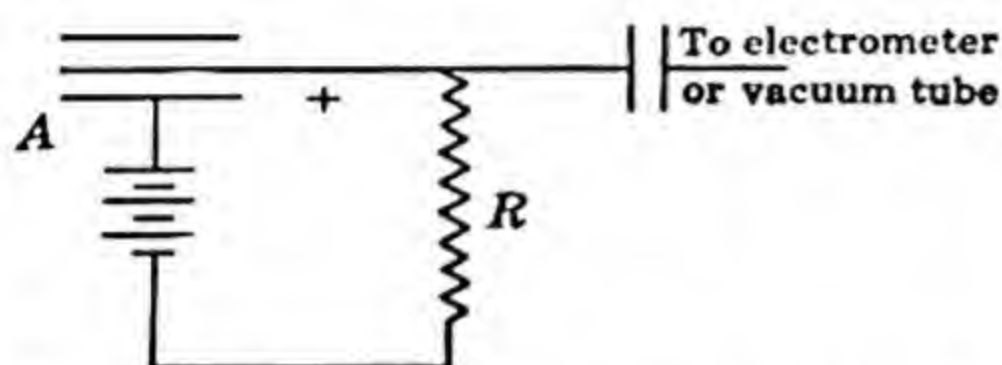


FIG. 6. Geiger counter circuit. There is a very high field between the cylinder and the wire of the counter *A*. An entering electron precipitates a discharge which causes current to flow in the high resistance *R* until the fall of potential across *R* reduces the potential across *A* to a point where the discharge extinguishes.

sufficient to cause a discharge. To make the counter sensitive again will now require the repetition of the charging and insulating process, which is obviously too elaborate. If some compromise between the dangerous maintained discharge and the self-ceasing discharge could be reached then the phenomenon of the discharge could be utilized in the detection of ionization.

In the Geiger-Müller counter a high resistance (about  $10^9$  ohms) is placed in the return to the high voltage as shown in Fig. 6. The entrance of an

electron then precipitates a discharge, but the resulting current must flow through *R* and in so doing sets up a fall of potential across *R* which greatly reduces the potential across the counter *A* and shortly extinguishes the discharge. The counter then gradually resumes its former condition of high tension and is ready to discharge on the arrival of a second electron. It can be seen that the high resistance fulfills the purpose of compromise as mentioned in the last paragraph and also ensures the automatic charging of the two electrodes so that the counter is in readiness for the next arrival. This automatic charging is not infinitely rapid, and a counter which employs a large series resistance will take a long time to recover after it has operated and so will only count slowly. This fact should be remembered when the operation of counters in conjunction with vacuum tubes is discussed a little later.

Since the potential of *A* is determined by the battery or power supply it is clear that the potential of the wire must change considerably in the above operation. In fact it changes by amounts varying be-



tween a few and a few hundred volts. The change in potential can easily be detected directly with a cathode-ray oscilloscope, or it may be converted into a current pulse by means of a vacuum-tube amplifier and detected by a counting circuit of the kind to be described later. Usually a single-stage amplifier can be used, which as often as not need not be shielded from electrical disturbances as the pulse supplied by the counter is so great that it appears very clearly above any disturbances.

The method of "quenching" the discharge is not always the same. Quite commonly a counter is filled with a mixture of gases, one of which is an organic vapor. This organic vapor is chosen so that it can absorb energy and *dissociate*. This process cuts down the number of photons in the discharge and acts to quench it. In time the organic vapor becomes used up, which is a disadvantage. About  $10^8$  counts can usually be made before this happens. In organic vapor counters the function of the resistance  $R$  is not quite the same and may actually be needed to maintain the discharge longer.

It is in place here to discuss the practical side of Geiger-counter construction. Prob-

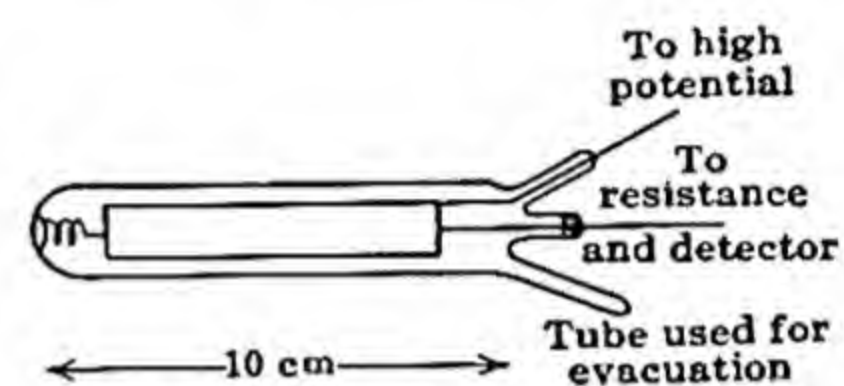


FIG. 7. Common design of Geiger counter.

ably no apparatus in modern physics has had more study from the technical side or appeared in more diverse successful forms than the Geiger counter. At the Massachusetts Institute of Technology the visitor is shown a Geiger counter consisting of a fork and spoon in a partially evacuated space. It works! On the other hand the reader may well intentionally make up a Geiger counter after the best instructions and fail to make it work. The authors would like to meet the counter expert who has not at some time in his career found he has constructed a "lemon." This is, however, too much of an aside. In practice the high field near a wire is almost universally employed in counter construction.

In Fig. 7 is shown the commonest form of a Geiger counter. A tungsten wire (from 1 to 10 mils) is stretched by a tungsten spring at one end and sealed along the axis of a glass tube which also contains a thin copper cylinder with a connector sealed into the glass. The glass envelope is attached to a pumping system after the contents have been thoroughly cleaned with soap and water and dilute nitric acid. The counter is then pumped out to a very low pressure with a diffusion pump system, and while on the pumps it is heated to outgas the metal surface. A little air can now be admitted and the heating continued



to oxidize the copper slightly; the counter is again pumped out and then filled with the gas mixture chosen. Argon and oxygen is one favorite recipe, about 8 to 1 in that order, at about 5 to 10 cm of mercury pressure. Self-quenching counters made of argon and alcohol in the same proportions are very satisfactory. It is important to avoid the use of any quenching compound which will react, however slowly, with the materials in the counter, for then the behavior of the counter changes with time. To set the counter in operation it is attached to

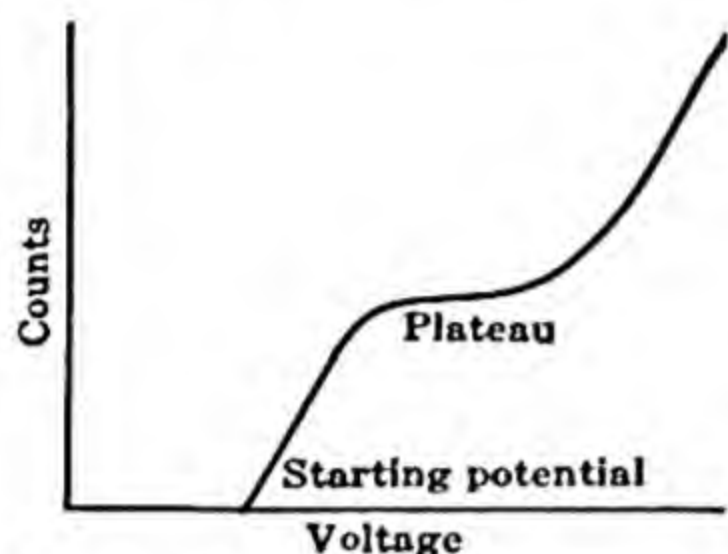


FIG. 8. Graph representing the number of counts observed when the voltage on a Geiger counter is increased. There is a definite starting potential, followed by a rise in the number counted, then a plateau, which means that as the voltage is increased there is very little change in the counting rate. After this there is a second rise.

a detection system and to a source of variable high potential. A source of radiation is then brought near, for example a piece of uranium glass or a luminous clock face, and the voltage is gradually increased until the counter begins to count. With counters about 2 cm in diameter and a wire of 10-mil tungsten, at a gas pressure of 8 cm argon and oxygen, the counting voltage is about a thousand. These figures are only approximate; in fact, this whole account of the construction of a Geiger counter is included more to give the reader an idea of the procedure involved than to act as a guide to actual construction. References for this are given at the end of the chapter.

If a counter is filled and sealed and made to operate it is very instructive to study the way in which the number of counts increases as the voltage on the counter is increased. Most counters behave as represented by Fig.

8. In this a graph of counts versus applied voltage is plotted for a counter exposed to a constant source of radiation. It can be seen that a certain starting voltage is needed before any counts at all are obtained, after which there is a steady increase in the number of counts until a more or less flat region, called the *plateau*, is reached. This plateau can be as broad as several hundred volts or may be entirely absent. Its extent depends on the value of the quenching resistance; it is generally larger when the quenching is done by vacuum-tube circuits. After the plateau there is a further rise, generally accompanied by a rise in the "background" count. This background count is due to cosmic rays and to spontaneous discharges, possibly caused by light. It is obviously advantageous to operate on the plateau, for then variations in the applied voltage are not important.



A most useful form of counter is shown in Fig. 9. It consists of a metal cylinder, closed at one end with a Kovar wire glass seal \* set in. A 5-mil tungsten wire is fastened to the insulated wire and the end of the wire carries a round glass bead. The end of the counter is closed by mica or aluminum. A side tube for filling is provided. Such a counter needs to be clean and free from dirt, particularly on the wire, and if filled with 8 cm of argon and 1 cm of alcohol will work practically every time at 800 volts with the Neher-Pickering quenching circuit. Under such conditions the counter has a short plateau of about 50 volts and operates excellently for counting beta rays.

Counters, recording equipment, and monitoring equipment are now available commercially. A very handy guide to this new "nucleonic" industry is the Radiation Instrument Catalog compiled by the Radiation Instrument Branch of the Atomic Energy Commission. This should be consulted by anyone interested in setting up a radiation laboratory.

The account just given of the operation of a Geiger counter assumes that everything is favorable. It would be leaving too rosy a picture if some of the difficulties were not mentioned. In the first place there is, particularly for the inexperienced, a considerable amount of trial and error

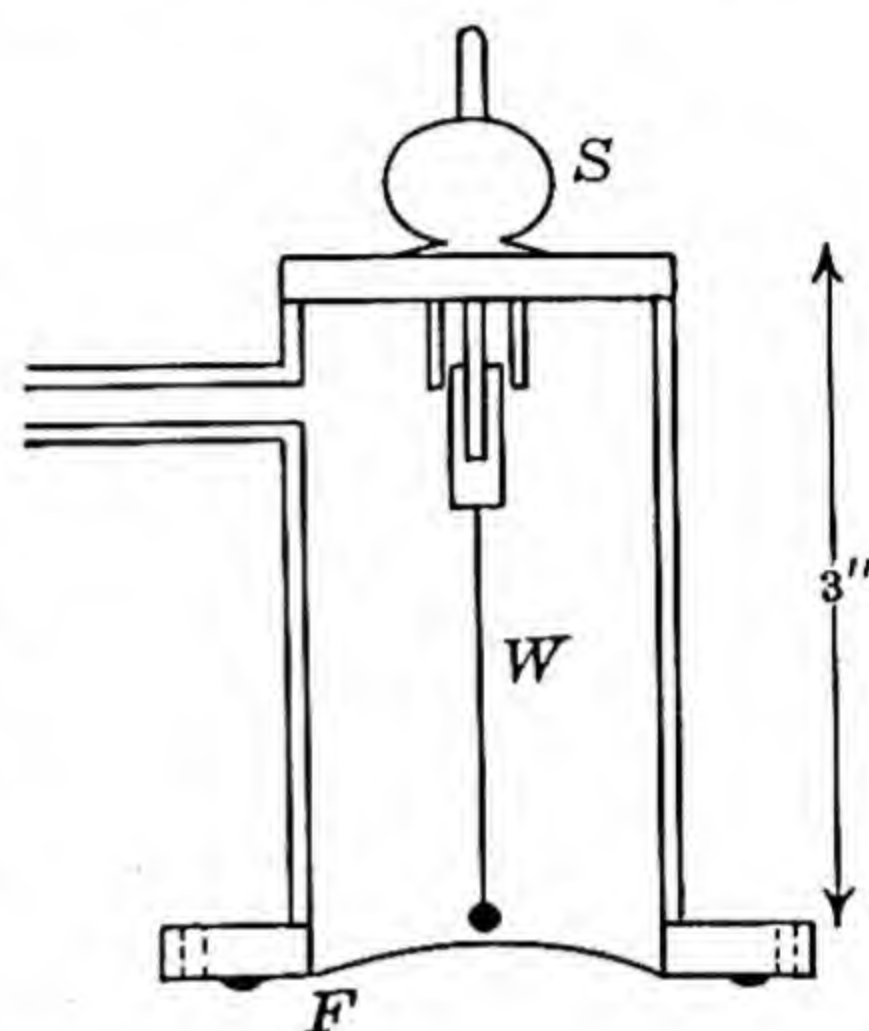


FIG. 9. Beta-ray counter with end opening. The wire *W* is of 5-mil tungsten with a small glass bead at the end. *S* is a metal-to-glass seal for insulation. A mica foil *F* is waxed onto the end with red sealing wax. Such a counter, used with a quenching circuit, has a plateau about 70 volts wide.

about counter construction. Of three similar counters, one may completely fail to operate for no clear reason. If, however, one thinks of the manner of operation of the counter this behavior is to be expected to some extent, for the counter must be such that it will discharge at a certain voltage when "tripped" by an entering electron and yet extinguish when the applied voltage falls by a hundred volts or so. Since the nature of the discharge in the counter depends greatly on the surface of the electrodes any dirt may or may not change the behavior of the counter in a radical way. Moreover, the gas used for filling plays an important role and must be chosen with care.

\* Available from the Stupakoff Co., Latrobe, Pa.



There is, therefore, in the author's opinion, no infallible set of rules which when followed will guarantee perfect operation of a counter; the proof of the pudding is emphatically in the eating. Preparation of Geiger counters is about on a par with cooking a meal; experience makes a difference. Once a counter is found which operates satisfactorily and does not contain reactive organic vapors and has no leak, it will continue to operate indefinitely.

In several laboratories as many as fifty counters are in operation at one time, giving no trouble, so that they are not uncontrollably temperamental.

The background count of a counter generally depends on the area of the electrode. About 1 per minute per square centimeter of surface is not a bad guess for most counters. It can be reduced by a factor of 2 by 2 inches of lead shielding.

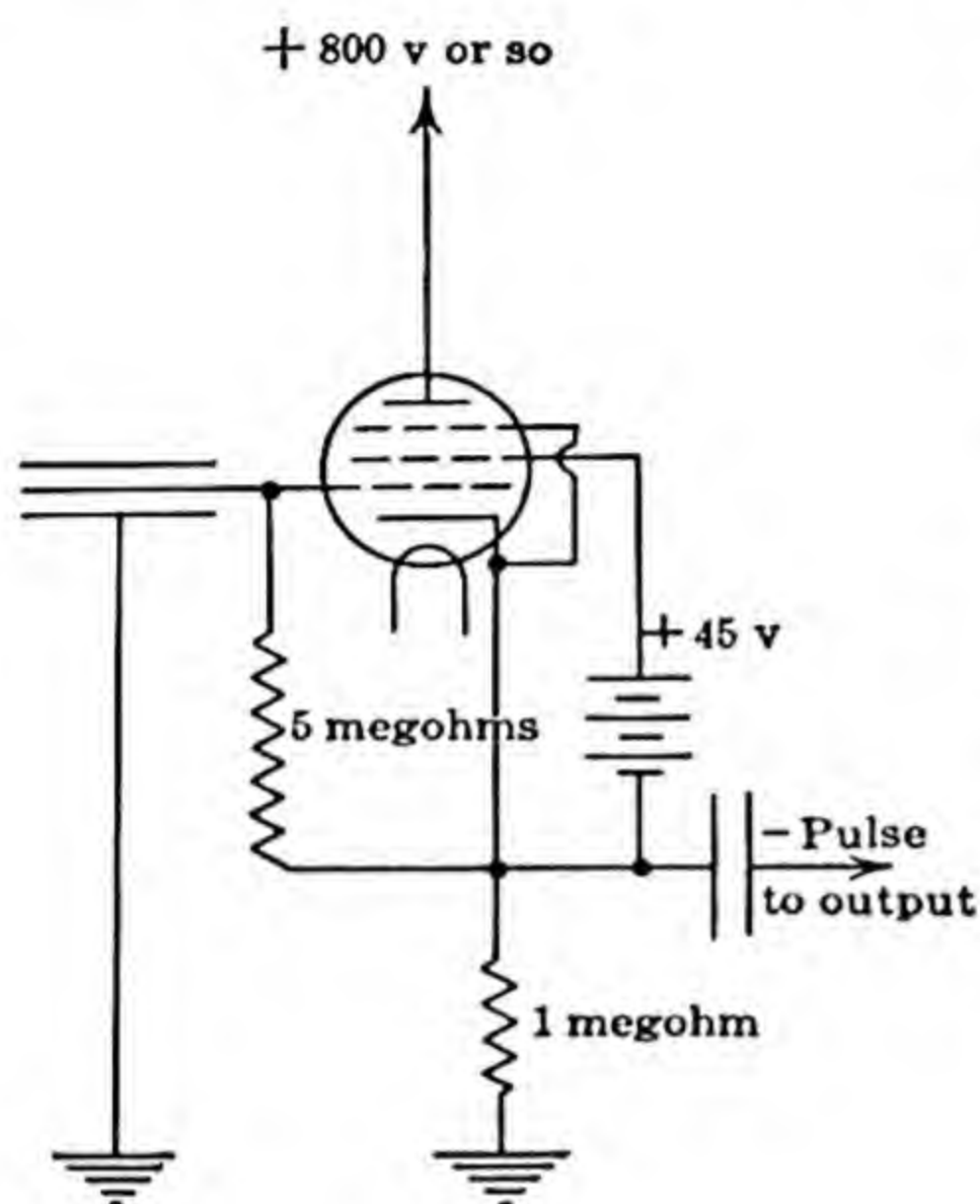


FIG. 10. Schematic quenching circuit. The pentode carries the high voltage to the counter wire. When the discharge starts the grid voltage downward, owing to the arrival of electrons the pentode ceases to pass current and the discharge is quenched. This is the Neher-Pickering circuit. It operates with the counter case grounded.

10. The high-gain pentode is in series with the high-voltage supply, and the arrival of electrons on the counter wire drives the tube to cutoff and stops the discharge. This circuit is due to Neher and Pickering and is called the Neher-Pickering circuit.

The fact that the Neher-Pickering circuit permits the cylinder to be grounded brings up an important technical point about counter operation in general. If the cylinder is not grounded the contact of

### Vacuum-tube control of Geiger counters

A very common device to quench a counter is to employ a vacuum tube. Many such devices exist. All utilize the fact that a vacuum tube can act as a rapid switch which can be actuated by the voltage change on the wire of the counter. For most purposes it is desirable to have the counter wire at high voltage and the cathode at ground. A switch which permits this and acts to break the high-voltage circuit is shown in Fig.



bodies at differing potentials with the glass case will induce potentials on the wire which may be sufficient to operate the recording mechanism. Thus, if a counter is connected so that the negative voltage is applied to the cylinder and a liquid is flowed around the counter, it will often be found that spurious counts will occur as the liquid is poured on, even after things should have settled down. If, however, the cylinder is at ground potential it acts to shield the wire and such spurious counts will be avoided. This is a small point, but the authors have known its neglect to waste much time.

### Scintillation counters

A curious turn of the wheel has brought scintillation counters into great prominence. Much of the early work in nuclear physics was done by the observation of zinc sulfide crystals containing a slight contaminant of copper. Such a technique had to produce enough light to be seen in a microscope, and the small number of quanta produced made counting very laborious indeed. The advent of the photo multiplier tube, which is sensitive to one photon, has changed this. Kallman discovered that naphthalene and anthracene crystals produce and transmit light which will excite a photocathode when ionizing radiations enter. Phenanthrene, sodium and potassium iodide, calcium tungstate, stilbene, and polystyrene are additional substances which are suitable for this purpose.

A scintillation counter using naphthalene is shown diagrammatically in Fig. 11. It is exceedingly simple to operate and, thanks to the availability of photomultiplier tubes all packaged ready for use, is perhaps the easiest counter of all to make work. The action is as follows: The charged particle, in ploughing through the naphthalene, ionizes many atoms and of course breaks many chemical bonds. A fair fraction of the recovery process after this devastation takes the form of the return of electrons to atoms with the attendant emission of light. For naphthalene this light is mostly of wavelength 3450 Å; for anthracene, 4440 Å. This light produces one or more electrons in the sensitive cathode of the photomultiplier. These electrons are accelerated by a voltage of about 80 volts to the next electrode whose surface is such that each 80-volt electron produces several slow electrons. These are in turn accelerated to the next electrode and so on until a considerable avalanche reaches the last electrode and a readily measurable current pulse is produced.



The great disadvantage of these counters is thermal "noise" which produces a background count of the order of thousands per minute. To reduce this the tube can be cooled with a block of dry ice. To some extent the great sensitivity of the counter makes up for this high background.

Such counters have four very solid advantages. First, they can operate in a vacuum, thus eliminating troublesome thin foil technique;

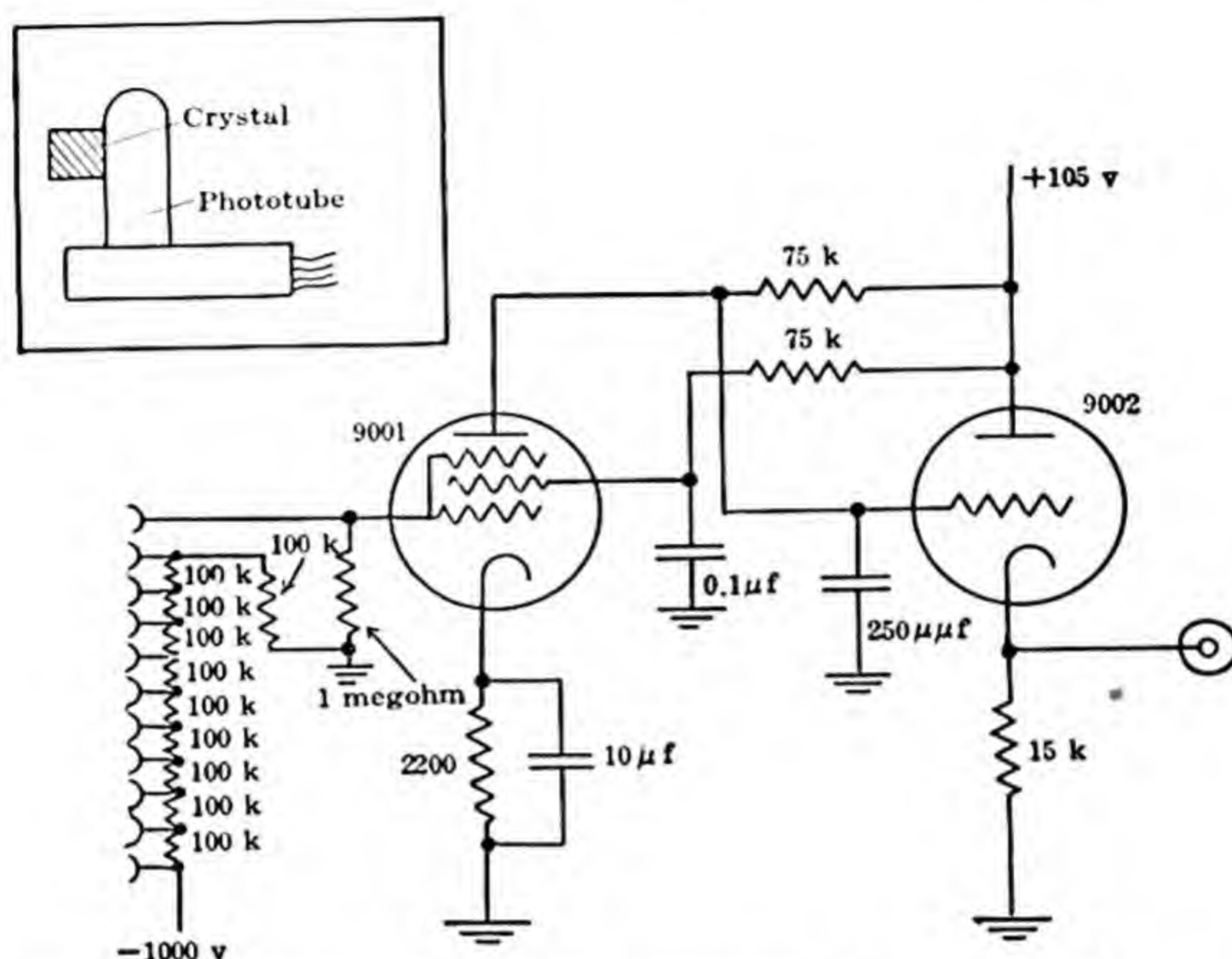


FIG. 11. Crystal, photomultiplier, and preamplifier circuit for a scintillation counter. The photo tube, shown in the insert, must be entirely shielded electrically by aluminum foil and from light by black friction tape. The background is reduced if the tube is held in a cardboard box filled with dry ice.

second, they deliver a pulse which is roughly proportional to the ionization entering, thus affording a means of distinguishing between kinds of radiation, something a Geiger counter cannot do; third, they can be made of considerable thickness and this, as will be seen later, renders them highly sensitive to gamma radiation; finally, such counters appear to act very rapidly so that response times of the order of  $2 \times 10^{-8}$  second are possible. It is therefore not surprising that much active research has gone into scintillation counting.

It is possible to reduce the background "noise" in the photomultiplier by using two phototubes to look at the same crystal. The noise pulses are random and not related while both tubes see the flash of light which accordingly actuates the coincidence circuit. This technique is elaborate but straightforward and is very promising.



## Recording equipment

Up to the present no word has been said about actual recording. For simple counting with relatively weak sources a thyatron recorder using a gas-filled 884 or 2050 tube is often employed. The thyatron is a three-electrode gas-filled tube which has a potential of 200 volts or so applied to its plate. If the grid is kept at a negative potential greater than roughly one-tenth of the plate potential there will be no discharge in the gas, or in other words the thyatron will not "fire." If, however, a positive pulse is put on the grid a discharge starts which is maintained by the power supply connected to the plate and which does not cease after the pulse has passed. The current passed by the thyatron depends on the nature of the tube and on the resistance in the plate circuit, but for the 884-type tube it is between 20 and 200 milliamperes in ordinary use. This is adequate to operate a high-impedance mechanical recorder or relay. The only problem is to extinguish the discharge in the thyatron after the pulse has been recorded. This cannot be done by the grid; it is necessary to bring the voltage drop between the plate of the thyatron and the cathode to less than that necessary to maintain the discharge, whereupon the tube ceases to fire and is once more under the control of the grid. One obvious way to cause this voltage drop is to make the recording mechanism break the plate circuit, and this can be done. It turns out to be rather unreliable, particularly for fast counting, and the method universally used is to short-circuit the tube by means of a condenser.

A simple circuit for this is shown in Fig. 12a. The pulse is applied through the input condenser to the grid of the thyatron, which fires. The act of firing causes the condenser  $C$  to discharge and the potential of the point  $P$  to fall below the voltage necessary to maintain the discharge. The thyatron then is extinguished and is ready for operation by a fresh pulse applied to the grid. It is possible to make the operation a little more certain if an inductance is inserted between the points  $A$  and  $B$ . This inductance, together with  $C$ , forms a series oscillatory circuit and so causes the potential of  $P$  to go *below* zero, thus making sure that the thyatron is extinguished. If it happens that the thyatron readily extinguishes but the recorder does not operate, a resistance of a few thousand ohms placed between  $A$  and  $B$  will slow down the fall of potential of  $P$  and permit the discharge to continue longer and so operate the mechanism.

The authors have found a rather more elaborate circuit suggested



by Schultz to be very satisfactory. The actual recorder is actuated by a 6V6 tube which provides the necessary current. The 6V6 is, however, controlled by a preceding circuit which produces a flat-topped pulse of variable width. This circuit is in turn triggered by

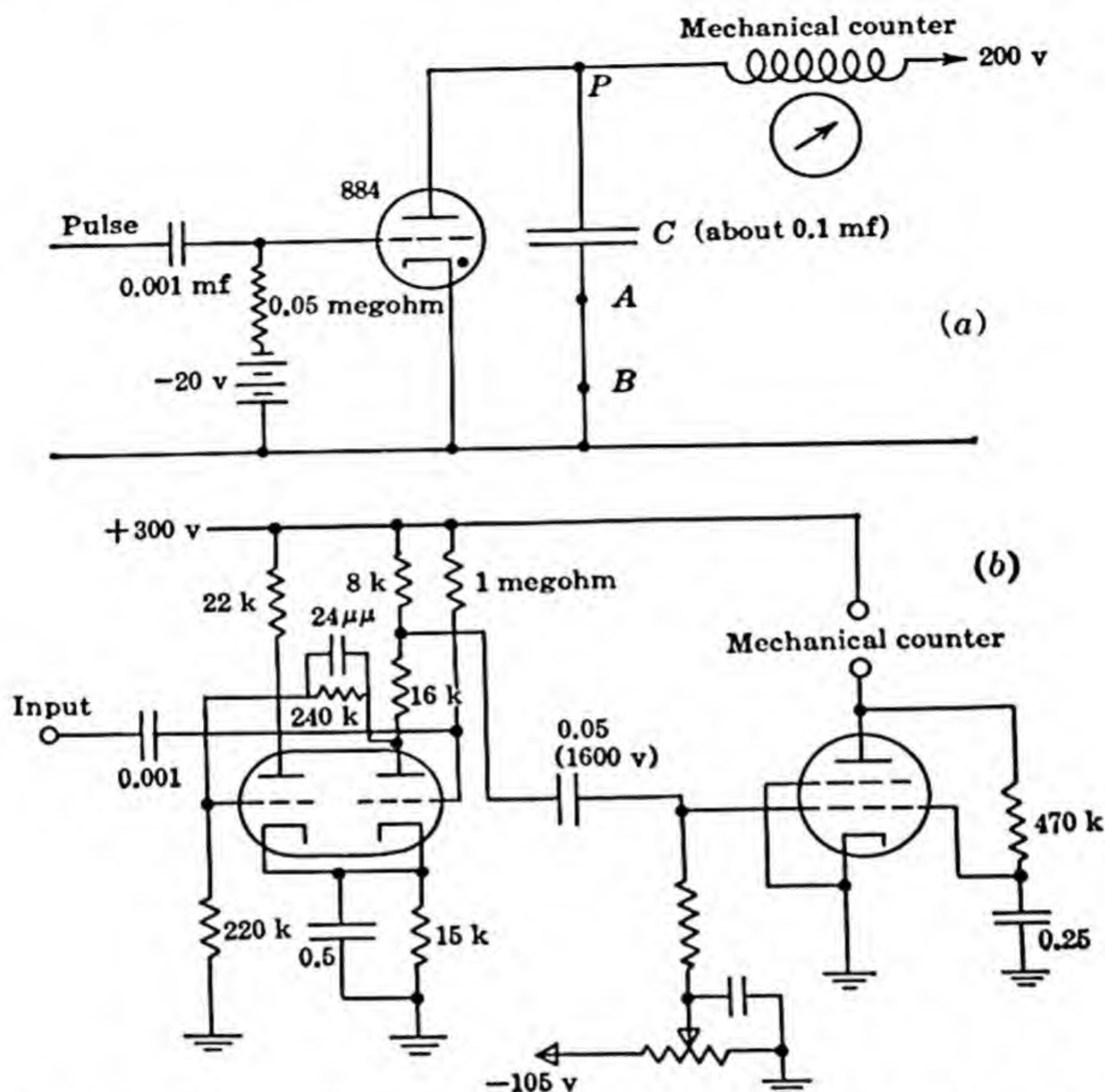


FIG. 12. Methods of actuating a mechanical counter. Fig. 12a shows a thyatron used for this purpose; Fig. 12b shows a more elaborate circuit in which the counter is operated by the current from a pentode which is turned on by a pulse delivered from a multivibrator circuit.

the pulse delivered by the particle counter. The whole circuit is shown in Fig. 12b. When a heavy counter such as a telephone message register has to be actuated a thyatron circuit as shown in Fig. 12a is necessary because the current required is much higher.

Such a recording circuit is almost instantaneous in action, and its speed is limited only by the mechanical counter. For speeds of counting up to 300 per minute this simple circuit is adequate. Beyond that, various so-called scaling circuits must be used of which the simplest, the "scale of two" circuit, is described below.



### Scaling circuits

The basis for many methods of handling large numbers of counts per second is the circuit shown in Fig. 13a. Because its operation involves only the control of the space charge in a vacuum tube as op-

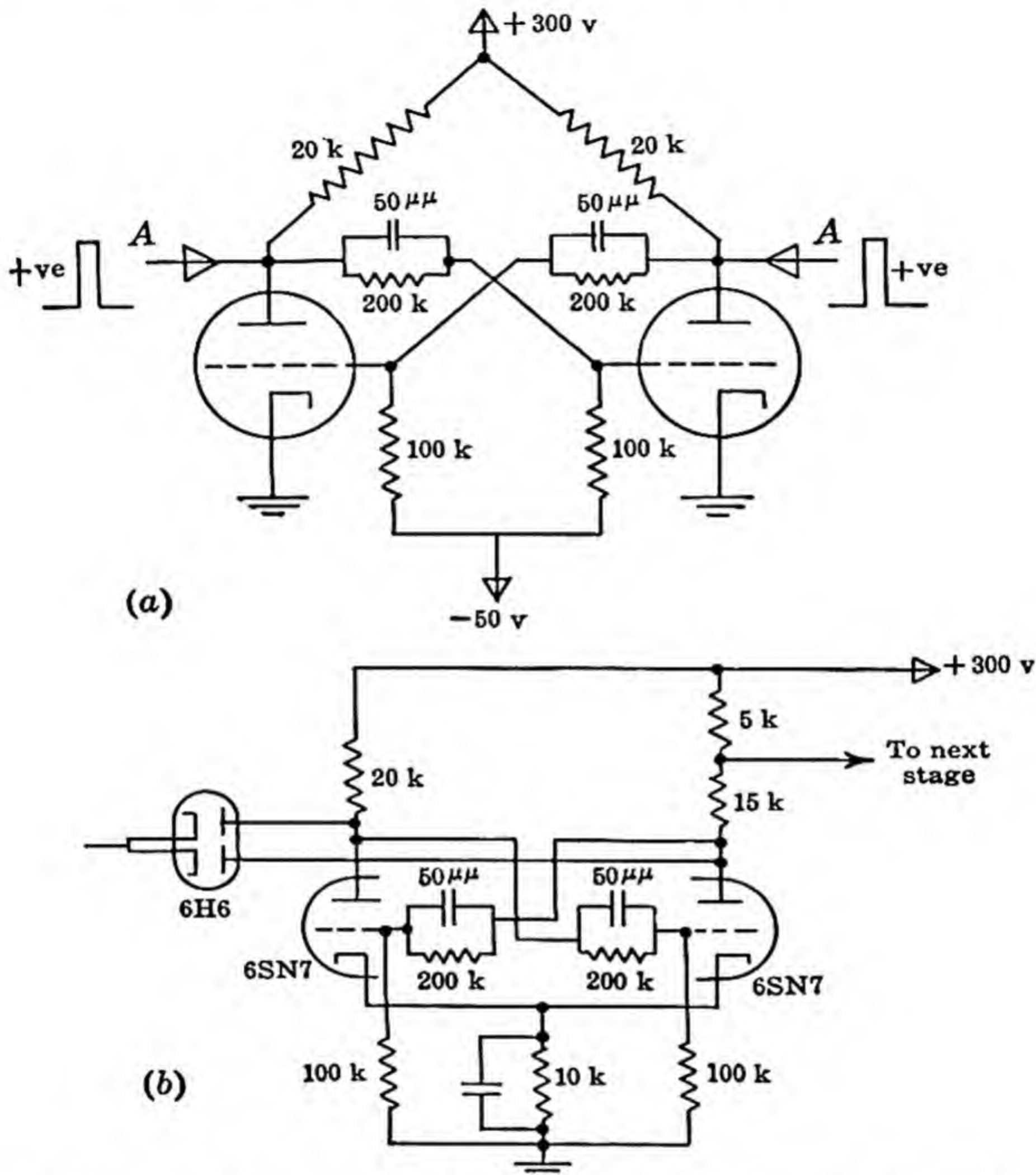


FIG. 13. "Scale of two" recorder. The circuit is statically the same for each tube but dynamically different in that the plate voltage of a tube which is conducting is lower than one which is not. The tube with higher plate voltage responds to the positive pulse applied to both. The current thus shifts from tube to tube at each applied pulse.

posed to a discharge in a thyatron its action is exceedingly fast. Counting rates up to 50,000 per second can be maintained with a series of these circuits.

The principal unit of the circuit is a symmetrical pair of triodes whose grids and plates are interconnected. Only one of these triodes



can be passing plate current because by so doing it holds the other one off. The circuit is symmetrical in a static sense but not in a dynamic sense. Thus suppose the first tube is on and equal positive pulses are applied at *A* as indicated. The plate voltage on the first tube is low whereas that on the second tube is much higher, being 300 volts. The positive voltage at *A* is communicated to both grids equally via the resistive and capacitative coupling, but the space charge in the second tube is much more responsive to the change in the grid voltage. Thus in the transient stage while the positive pulse is applied at *A* the off tube responds more quickly, and as a result the tube current shifts from the first to the second. Thus if an output pulse is taken from either tube it will occur every other time. This accounts for the name "scale of two." Because a scaling circuit depends on this dynamic unbalance it is quite important that the biases and resistance values be correctly matched to the tubes.

Very rapid counting indeed is practicable except that the random nature of nuclear processes makes very short intervals between pulses quite probable. This means that the first two or three units of a scaling circuit must be able to change rapidly, requiring small plate resistances and a fair amount of power. The later stages are not at all critical.

Usually the pulses are fed in through a buffer stage. Double diodes and triodes lend themselves handily to this process and very compact scaling units can be constructed. The circuit for one such unit, designed by Higinbotham, is shown in Fig. 13b.

### Counting-rate meters

For some purposes it is convenient to use a counting-rate meter in place of a scaling circuit. Such a meter simply reads the *rate* of arrival of the pulses, and if this rate is rapid enough the fluctuations are not very great so that a reasonably steady reading can be obtained. Such an arrangement is most convenient for photographic recording. A very carefully designed meter has been described by Gingrich, Evans, and Edgerton and tested very thoroughly by them. Their circuit is carefully arranged to provide pulses of the same size so that each count records in the same manner. These pulses are then fed into the grid of a pentode which is biased to give very little plate current. The pulses produce little bursts of plate current which can be read on a meter. The readings so obtained are very unsteady, but by placing a 100,000-ohm resistance in series with the meter and a 20-microfarad condenser across the two the fluctuations can be smoothed



out and a reasonably steady deflection results. Time must be allowed for the condenser to become charged, which means that the meter is not so direct as might at first be supposed, but it is still more convenient than stopwatch counting. If desired, the small steady plate current can be balanced out by a potentiometer arrangement in the plate circuit. The meter must, of course, be calibrated.

A very rapid counting-rate meter has recently been designed by Schultz. The counting-rate method is perhaps the best of all methods for counting at high rates.

Counting circuits of all kinds are available commercially, and one remarkable development has been the nucleonics industry, which has arisen to meet the new demand very rapidly and effectively.

### Fluctuations in counting

We have spoken above of the fluctuations in counting nuclear particles, and as they constitute a phenomenon which is inherent in all nuclear observations it is important to appreciate the nature of the random fluctuations which are met in this work. In radioactivity we may speak of certain definite figures which express the rate of decay of a substance, generally its "half-life." Such figures refer to measurements which involve the averaging of a very large number of observations of individual transitions. The actual process of the conversion of one atom of, say, radiosodium into stable magnesium takes place *at random*. It is pure chance when the individual event takes place. A very large number of such events, when averaged, will show a clear and accurate manner of decay, but the fact remains that the random nature of the individual process underlies the whole general phenomenon.

This shows up very clearly when a weak source of a radioactive substance is placed near a counting apparatus and the number of registrations in every 10-second interval is recorded. One might well record in ten such intervals the numbers 8, 5, 6, 4, 10, 7, 6, 6, 7, 3. The average is 6.2, but as high as 10 and as low as 3 can be recorded. The arrival of the "counts" is much the same as the arrival of raindrops on a window pane. Such random fluctuations are encountered throughout atomic physics; for example, "tube noise" is due to the fluctuations in the motions of electrons in a vacuum tube, and the finite width of a spectral line is due to a similar cause. It is only in nuclear physics that the individual particles are sufficiently energetic to be individually detectable and the fluctuations show up directly



instead of as a secondary effect concerned with the measuring apparatus.

Since these fluctuations are inevitable, what must one know about them? In the first place one must be wary of data taken with only a few counts. In the second place one must be prepared to accept limitations imposed on experiments by the method of counting available. In the third place one should know rough limits of error to any count which is taken.

There is one very simple and complete way of considering any one observation. This is by means of Poisson's formula. If, in a given interval, a number of particles  $n$  is counted, and if the average number arriving in a large number of such intervals is  $x$ , then the probability of the occurrence of  $n$  is given by

$$P_n = \frac{x^n}{n!} e^{-x}$$

where  $P_n$  is the ratio of the number of times  $n$  would be recorded, to the whole number of trials, if a very large number of trials were made. Now in analyzing data we wish to assume that  $n$  as observed represents the true average  $x$ . *We have no guarantee that it does*; that is the inherent uncertainty with which we must be content to live. We can, however, suppose that  $n$  is in error by a certain amount, say the permissible limit of error, and can then calculate the probability that this is so. If the probability is low we can be satisfied; if it is of the order of 10 per cent it is necessary to worry. To illustrate this method of discussion we give the probability of various errors in a count of 1000 particles. The probability is expressed as the ratio of the probability of the observed 1000 to that of the true average.\*

TABLE 1

POSSIBLE "TRUE" COUNT	RATIO OF PROBABILITY OF THE OBSERVED 1000
1010	0.9
1020	0.8
1050	0.3
1100	0.03
1125	0.01

It can be seen that a 5 per cent error is quite likely and that there is a definite chance of an 11 per cent error. In the great majority of

\* This means that, if a large number of trials were made, and the average were 1010, the count of 1000 would be obtained 0.9 times as often as the count of 1010 and if 1125 were the average, 1000 would occur 0.01 times as often as 1125.



experiments an elaborate discussion of the errors due to fluctuations is not presented, but the numbers counted and the conclusions drawn are such that there is an ample safety factor. As a rough guide one can expect a fluctuation as great as the square root of the number of particles counted in any one observation. This guide is useful because it indicates the improvement to be expected if more care is taken. Thus a count of 100 particles is fairly reliable to 10 per cent, but nearly 1000 must be counted to be fairly sure to 5 per cent. A habit the authors have formed when there is a little question of the validity of the conclusions drawn is to divide the observations into two, taking the first half of the data and comparing it with the second. If the two agree it is reassuring. By far the best procedure if there is any question regarding fluctuations is to take more data.

### Coincidence counting

Nuclear processes generally take place in less than  $10^{-13}$  second. Therefore if it is possible to observe separately two events connected with the same nuclear process the fact that they are connected shows as a *time coincidence*. This time coincidence is very precise unless a reason for a delay exists. When this delay exists it is usually interesting to make a special study of it.

The simplest illustration of a coincidence is that of a fast particle traversing two counters and recording in each. If one counter is placed above a cloud chamber and a second below, then a time coincidence between the two means that a particle has traversed the space between them and therefore also the cloud chamber. If the coincidence is made to actuate the expansion of the cloud chamber the ionization left by the particle is still there and therefore the track of the particle can be seen. This method, introduced by Blackett and Occhialini, has revolutionized cosmic ray research.

Less obvious but equally important is the time coincidence existing between gamma radiation and other processes such as the emission of a beta particle or a proton in a transmutation. The study of such coincidences is often the only way to decide on a scheme of energy levels. Occasionally delayed coincidences are observed, corresponding to a transition process of short average duration, the most famous being the decay of the  $\mu$  meson to an electron as observed by Rasetti.

The most important factor in coincidence counting is the possibility of random coincidence. If the counter or the circuit has a finite time of response  $T$  (called the resolving time) and the two counts are  $N_1, N_2$  particles per second, then there will be random coincidences



at a rate  $2TN_1N_2$  per second. This is easy to see. Consider the events in 1 second. The first counter will actuate the circuit for a total time  $TN_1$ . However, the first counter will take a share in a coincidence with the second counter for double this time because the second counter has only to operate the circuit for a fraction of the overlapping time. This total overlapping time is  $2T$  per count in the

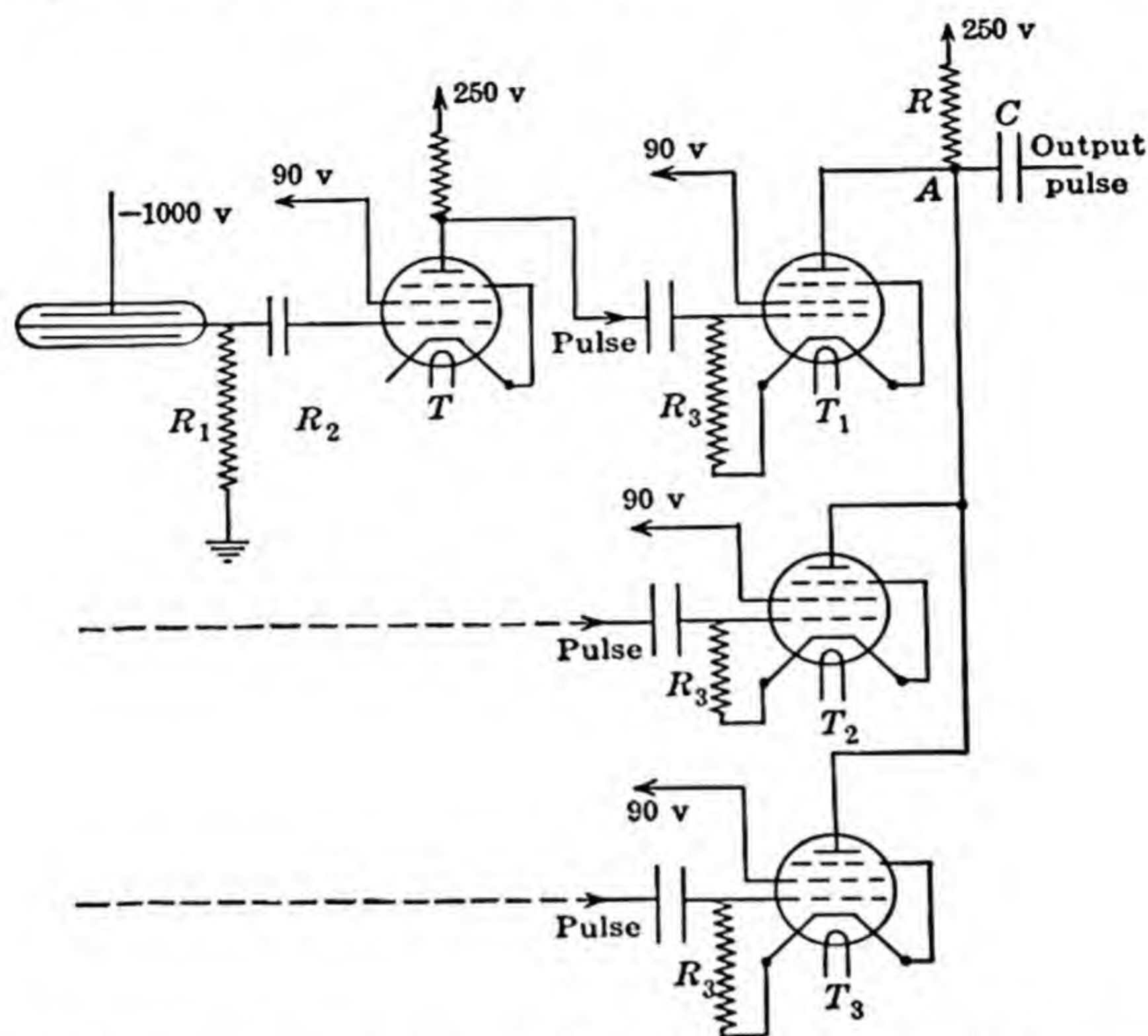


FIG. 14. Triple coincidence circuit. The Geiger counters deliver pulses to the three tubes  $T_1$ ,  $T_2$ ,  $T_3$ , such that their grids become negative and plate current ceases to flow in the resistance  $R$ . When this is so the potential of  $A$  rises and an output pulse is delivered. If any *one* tube is not blocked, enough current flows in  $R$  to keep the potential of  $A$  low, and hence a pulse is not delivered unless all three counters operate simultaneously.

first counter.\* Now we can regard each count in the second as able by chance to occur in the time  $2TN_1$  pre-empted by the first. The fraction of such counts causing a coincidence is thus  $2TN_1/1$ , the ratio of the "active" time to the total. The number of attempts made at such occurrence by the second counter is  $N_2$  so that the total number of successes is  $2TN_1N_2$  per second.

\*If the reader prefers to visualize this we suggest he draw two pedestals of width  $T$  on two lines and then gradually draw them apart. At the limiting contact the conditions are as though there were one pedestal of width  $2T$  making coincidence with a line. It is this limiting idea which is used above.



This expression for random coincidences is the dark side of life for the coincidence expert. It represents meaningless data. To reduce the number one can reduce  $N_1$  and  $N_2$ , but that lengthens the experiment. It is better to reduce  $T$ , and modern circuits can employ a value of  $T$  which is under  $10^{-7}$  second.

The other factor in coincidence counting is efficiency of detection. This must be as high as possible or else the ratio of true to random coincidences is not high enough. In cosmic ray research where  $N_1$  and  $N_2$  are necessarily low, coincidence counting has enjoyed its greatest success. Much current work in nuclear problems is now going forward, aided by scintillation counters.

Two types of coincidence circuits are in common use. For multiple coincidences the Rossi circuit shown in Fig. 14 is used. The principle of this lies in the fact that if all tubes sharing a common plate resistance are simultaneously turned off the voltage at the plates rises to the voltage of the supply, whereas if even one is still on the plate voltage stays quite low. A multiple coincidence of all the counters thus produces this large voltage change. A second circuit has been developed by Schultz. This operates on the principle that if two sharp triangular pulses are superposed the total voltage can only reach double of each when exact coincidence occurs. Thus by means of a bias a variable resolution time can be achieved. By the use of wide band circuitry including special pulse transformers the resolving time can be reduced to less than  $10^{-8}$  second. With this circuit the time of flight of a cyclotron beam through a path 1 meter long has been observed.

Simple modification renders the Rossi circuit an *anticoincidence circuit*. The circuit then rejects time coincident events. This is often useful. For example, to ensure that a particle has stopped in a cloud chamber, a counter placed below in anticoincidence rules out all those particles that traverse the chamber. The cloud chamber is operated only when the lower counter is not affected.

## The cloud chamber

The cloud chamber is almost certainly the most widely known method of detection of nuclear particles because it gives such a graphic portrayal of the path of a nuclear projectile. The familiar principle depends on the observation of C. T. R. Wilson that ions can act as centers for the formation of condensed droplets in a supersaturated vapor. He produced the supersaturated vapor by the simple process of rapid adiabatic expansion of a saturated vapor. The rapid expan-



sion causes a fall in temperature which carries the vapor below the dew point. If the vapor is clean, particularly if it is free from dust, the state of supersaturation is maintained unless some ions are present. Droplets form about these ions and so make the presence of the ions known. By illuminating the vapor after expansion, photographs can be taken and any interesting events permanently recorded.

The cloud chamber is one of the oldest instruments in nuclear physics. It has undergone considerable development and has probably taxed more people's patience than any other piece of equipment in the whole subject. If the reader thinks for a moment of the method of operation outlined above he will soon see one disadvantage of the instrument. This is the fact that time must elapse before the cloud chamber is ready for a second expansion. The time of the expansion itself is short, and the sensitive time is about  $\frac{1}{30}$  second, but 20 seconds must elapse between expansions. This means that the apparatus is only  $\frac{1}{6}$  per cent efficient, and in turn this means long hours of work to secure data. Many attempts have been made to develop a "continuous" cloud chamber, and some success has been attained in finding means to produce a supersaturated state by a steady flow of one gas against a vapor. No research results have been produced yet by such equipment. A more useful technique has been to keep the cloud chamber in readiness for expansion until a particle is known to have passed through it. This seeming miracle can be accomplished only if the particle to be detected is an energetic one of the kind to be found in cosmic rays, a particle which can pass through the walls of the cloud chamber and also through a Geiger counter. The operation of this counter trips a relay which sets off the expansion. By this means the efficiency of the expansion chamber is enormously increased and the saving in film is great.

A cloud chamber for observation of alpha particles, protons, and heavy recoil atoms is not hard to construct. The expansion can be made in a volume enclosed by a rubber diaphragm which is compressed and then released. In place of water as the vapor, various alcohols can be used. They have the advantage that the "expansion ratio," the ratio of the volume after expansion to that before, is less. For air and water the expansion ratio is about 1.4, whereas with various alcohol mixtures it is around 1.2. The expansion ratio can be calculated in terms of the specific heats of the gas and latent heat of the vapor concerned. For a gas like acetylene with water vapor it rises to 1.8. The observation of fast electrons is much harder. The expansion must be kept within close limits defined by no tracks visible and all tracks



obscured by too dense a general fog. Any small trace of acid fumes will effectively prevent the operation at all, and if one talks to workers in cosmic rays one becomes aware of the existence of a cloud chamber "season," namely, the time of year when it is not too warm and humid. The observation of fast electrons by means of cloud chambers should not, therefore, be attempted lightly.

Expansion chambers can be operated at high pressures, up to 10 atmospheres. Such chambers are useful in measuring the ranges of protons emitted in nuclear reactions and in detecting the presence of groups. The recoil protons caused by neutrons in a gas containing hydrogen can also be detected with such equipment.

### Photographic detection

Photographic detection has always had considerable appeal because it offers the same advantages as the cloud chamber, namely, the ability to see the process occurring and to record it permanently. It was first used by Mariette Blau, and recently as a result of the efforts of T. R. Wilkins and C. F. Powell emulsions of sufficient thickness and fineness of grain have been produced for a wide range of use. The most successful of these to date has been marketed by Ilford of England. Emulsions up to 500 microns in thickness have been produced. The usual thickness employed is 100 microns. Such emulsions must be developed with great care, each process taking several hours, and they have to be handled carefully to avoid peeling.

The tracks of nuclear particles appear in a manner much like those of cloud chamber pictures. The ionizing particle causes the dislodgement of electrons in the emulsion. These electrons are held in position so that a latent image is formed which remains for a period of weeks or months. Development by a reducing agent then causes each grain containing the dislodged electron to blacken. Since the total path length of a nuclear particle in the emulsion is very short the grain size must be small or no kind of a track is produced. This technical difficulty has now been overcome. Figure 15, due to Dr. R. A. Peck, shows tracks of protons in photographic emulsion. They are easily recognized and readily counted.

The tracks are observed with a high-powered binocular microscope which needs to be focused at different depths in the emulsion. It is the tediousness of this work which detracts from the photographic method. This can be overcome by a proper psychological approach, which consists in observing in a group, rather than alone, and in an undarkened room. Morale rises each time an exciting process is un-



covered, and competition stimulates discovery while providing healthy skepticism. The group at Bristol in England presided over by Mrs. C. F. Powell operates in this manner, and their output of discovery has been amazing.

The undeveloped images on nuclear emulsions fade in about three months. Ilford plates can be operated in vacuum.

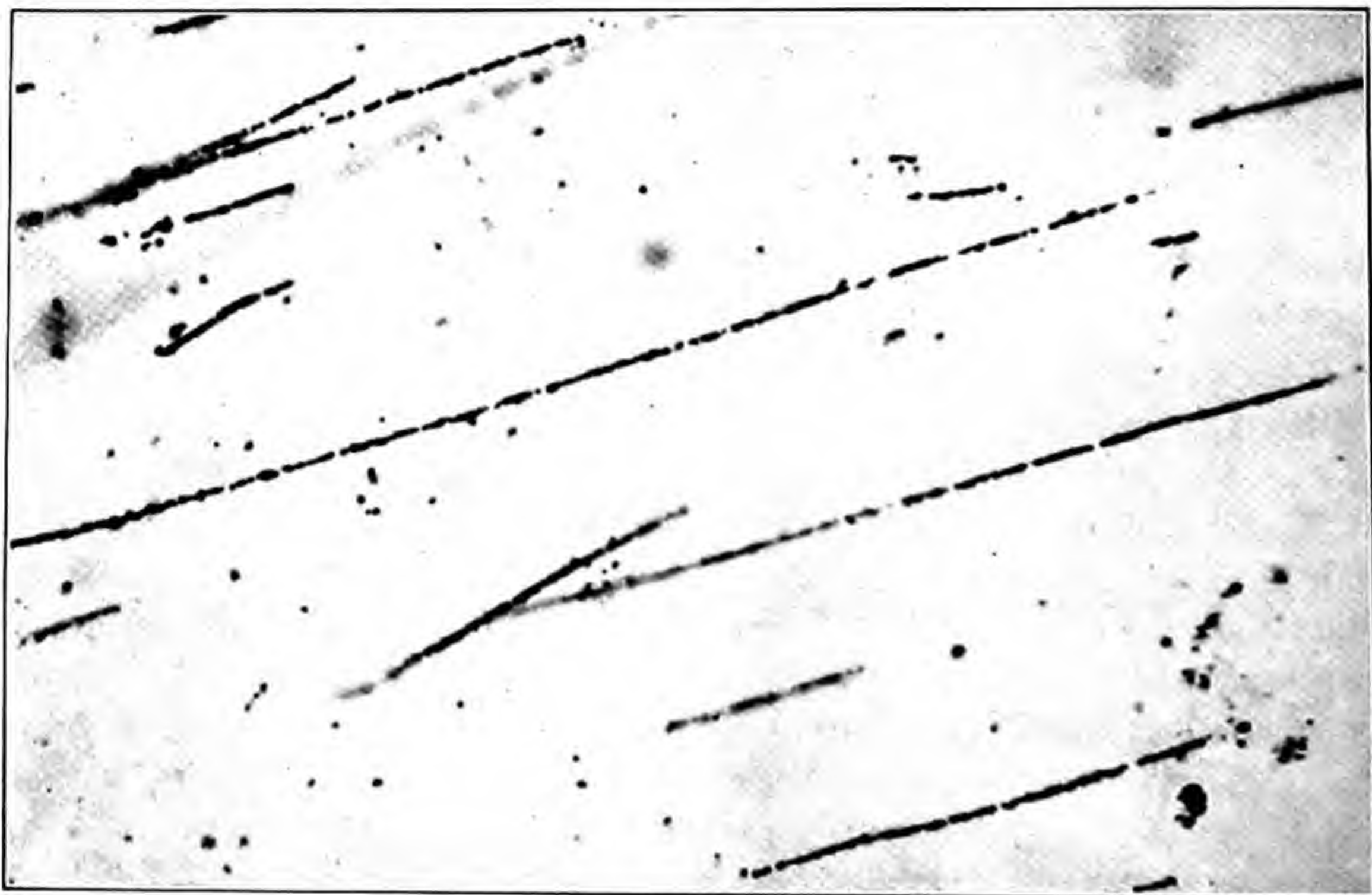


FIG. 15. Tracks of protons produced in the bombardment of phosphorus by deuterons as recorded in a photographic emulsion. The magnification is  $1150\times$ . (Record by Dr. R. A. Peck.)

Photographic technique offers the unmatched advantage of recording the total ionization of a particle as well as the rate of ionization. This has made possible the discovery of the various kinds of mesons and is being exploited in observing their properties.

An important improvement has been made by Kodak, Ltd., of England which makes it possible to observe light electrons. With such an emulsion the decay of a meson into an electron has been photographed.

### **Amplification of ionization current: the electrometer tube**

Where fairly large samples of elements having long half-life are to be studied, or where it is important for health hazard reasons to have a permanent record, it is often desirable to record the ionization cur-



rent produced automatically. A counting rate meter achieves this but is susceptible of circuit instability unless carefully constructed. Another method is to amplify the ionization current in an ionization chamber by means of a vacuum tube, often called an electrometer tube. Since the output of such a circuit can be made to operate a galvanometer, a permanent record can be produced.

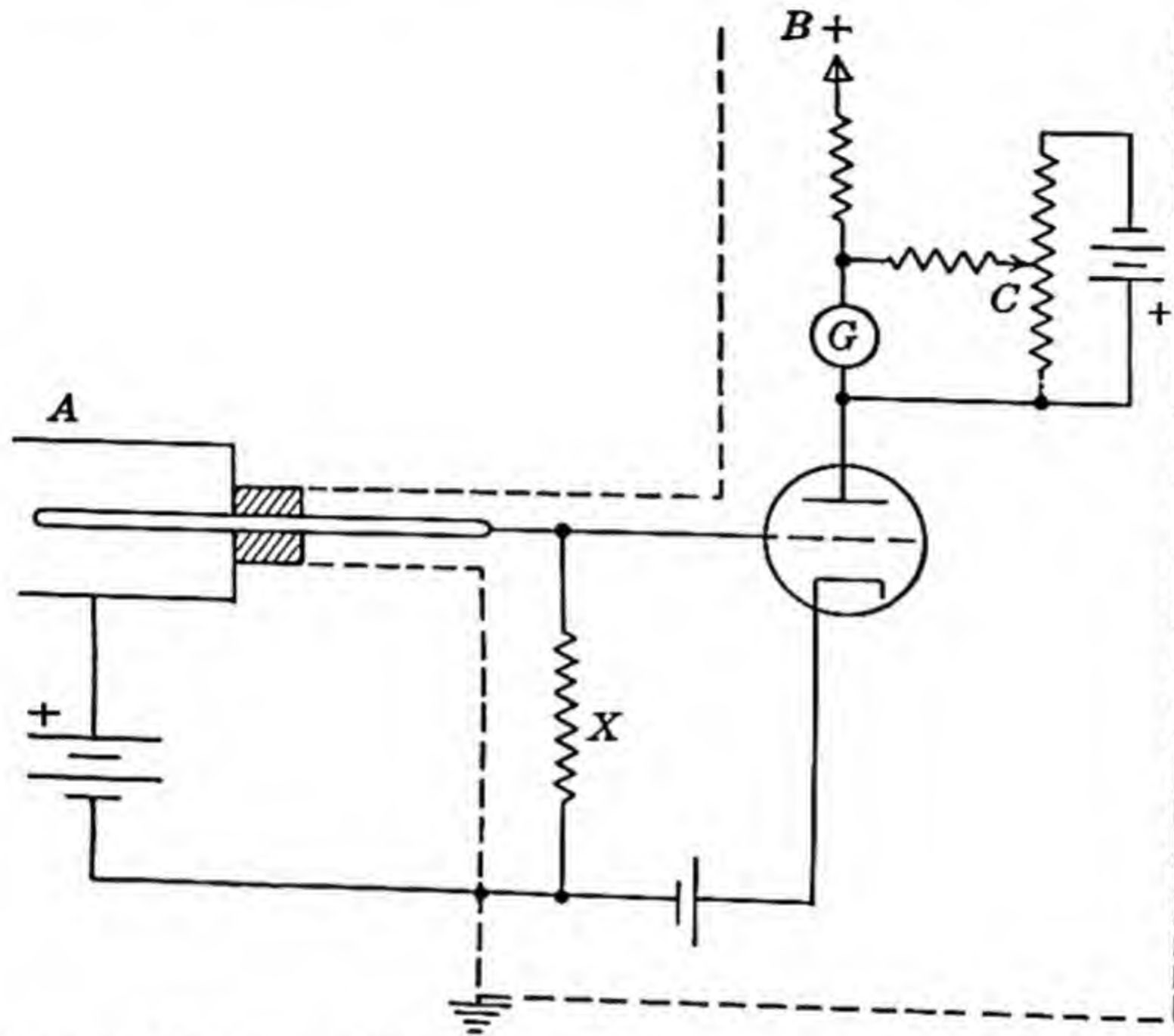


FIG. 16. Electrometer tube. When no ionization occurs in A only grid current flows in the resistance X and a steady grid potential results. The plate current is steady. The galvanometer G is brought to zero by the balancing circuit C. When ions are formed in A, additional current flows in X, changing the grid potential and the plate current. This change shows on the galvanometer.

The circuit of Fig. 16 shows how such an electrometer tube operates. Suppose that we consider the behavior of the vacuum tube if no ionization occurs in the ionization chamber. The plate current will be determined by the grid potential, which in turn will be determined by the bias battery and the flow of grid current through the resistance X. A steady state will be reached in which a steady plate current will flow, and this plate current can be read by a galvanometer, or a battery and variable resistance can be arranged to oppose the plate current so that the galvanometer shows no deflection in this steady state. Now, if ionization occurs in A, current flows through X and the potential of the grid is changed, thus causing a change in plate current. It is commonplace for a vacuum tube to produce a milliamperere of



plate current for a volt change on the grid, so that a change of 0.0001 volt would produce a current change of  $10^{-7}$  ampere in the plate current; a relatively insensitive galvanometer will readily detect this amount. If the resistance  $X$  is  $10^{11}$  ohms the current to produce the 0.001-volt change would be  $10^{-14}$  ampere, or about 33,000 ion pairs per second. This much ionization is produced by one alpha particle per second or by 100 electrons per second. As quite small sources emit at these rates, the method of detection is amply sensitive.

The catch appears as soon as the apparatus is set up. Not only the ionization chamber, but also variations in filament temperature, plate supply voltage, electrical disturbances, and so on, will produce changes in plate current as great as those given above. It is therefore necessary to use careful shielding, a large storage battery on the filament, good heat insulation, and a steady source of plate voltage before these sensitivities can be approached. By using multi-grid tubes, compensating networks can be devised to alleviate some of these sources of disturbance, and in competent hands the electrometer tube is one of the most satisfactory methods of observation of nuclear processes. It has one interesting application in which an ionization chamber at 10 or so atmospheres is used. The increased pressure means that a large part of the ionizing path of a beta ray is spent in the chamber and the ionization current is accordingly greater. Such an arrangement will detect single electrons and is probably the best possible method of detecting very weak sources.

The disadvantage of a sensitive galvanometer can be avoided by inserting a "chopper" at an early stage in the circuit. This interrupts the circuit at a rapid rate, say 40 cycles per second and permits amplification by an alternating-current amplifier tuned by special circuitry to the right frequency. Sufficient stable amplification followed by rectification can be obtained in this way to operate a relatively rugged meter.

### **The detection of neutrons and gamma rays**

We have made the point very firmly that charge and speed are needed for the detection of nuclear particles. What then is done about neutrons and gamma rays which possess no charge and (for neutrons) may have very little speed? The answer is that the detection process is *indirect*: first a secondary effect must be produced. This secondary effect then has the necessary charge and speed.

It is fairly easy to see how neutrons are detected. They interact readily with all nuclei either by billiard-ball type of collisions or by



being captured and producing nuclear changes. Skill in detecting neutrons consists in isolating one of these effects so that it may be used quantitatively. Suppose the billiard-ball process is chosen. One could pick hydrogen as the target ball. The neutron then collides with it and projects it forward. The hydrogen which is now moving rapidly sheds its electron and becomes charged and so can be detected, for example, in a proportional counter, or in a photographic emulsion. Other methods of neutron detection involve either the production of radioactive nuclei which emit beta particles and so are detected, or the splitting of light or heavy atoms into two parts which have charge and speed and so can be observed. These various processes will be considered later.

Gamma rays act somewhat differently. Predominantly a gamma ray reacts with electrons. These are either produced when the whole gamma ray is absorbed, thus producing photoelectrons, or when a collision occurs, producing "Compton" electrons. In both cases the electron is now the secondary product and has the qualities of charge and speed requisite for detection.

Common to both these processes of detection is the question of *transition* which we can briefly discuss. In general neutrons and gamma rays react infrequently with atomic particles. This means that lots of such particles must be available for interaction, a long-winded way of saying that the substance responsible for the secondary process must be thick. This very thickness, however, must be watched, for the secondary particles can ionize and so can be absorbed rapidly. This means that only the exit end of the thick layer does us any good in detection because every other region gives rise to electrons or protons which are absorbed before they reach the counter. This problem of transition greatly reduces the efficiency of detection of neutrons and gamma rays.

Simple considerations show that without tricks the detection efficiency is of the order of 1 per cent. As shown in Chapter 2, an electron of energy 1 Mev has a range of 383 milligrams per square centimeter of aluminum. This is about a millimeter and a half actual thickness. It is therefore no good to use material for conversion of 1-Mev gamma rays to electrons (often called the *radiator*) of thickness in excess of a millimeter and a half. In Appendix 5 it is shown that the absorption coefficient for gamma rays of 1 Mev in aluminum is 0.16 ratio per centimeter, meaning that in 1 centimeter of aluminum about 16 per cent of the gamma rays are absorbed. Only  $0.15 \times 16$



or 2.4 per cent are absorbed in a millimeter and a half. This is all the gamma radiation which can be detected. The actual figures are lower because very few of the secondary electrons have the full energy of 1 Mev.

For neutrons the situation is worse because the secondary products are more easily absorbed and the neutrons themselves are much more penetrating.

This obstacle of transition has stimulated methods of overcoming it. For gamma rays the scintillation counter in which the secondary electron is made to produce photons which in turn act on the photocathode is a great advance. The photons traverse the transparent naphthalene with ease so that the thickness of naphthalene can be as much as an inch. The efficiency is accordingly nearer 10 per cent. For neutrons the trick used is to slow them down so that their penetrating power is greatly reduced, and then let them produce a change in some nucleus which can be detected. Since both these require further knowledge of nuclear processes they will be deferred until later.

### Summary

To summarize, we can say shortly that to detect nuclear particles we detect *ionization*. This can be done directly by electrometers whose sensitivity can be pushed with difficulty to detect single particles. If 100 times the ionization of a single particle per minute is to be detected it can be done directly with the Lauritsen electroscope, which is about the most useful general-purpose instrument in radioactive work available. By means of tube amplifiers either single particles (high-gain amplifier) or steady ionization currents (single electrometer tube) can be observed. The latter is very well adapted to be used with a recorder. Use of a high electrostatic field and ionization by collision renders the detection of single electrons easy. The most widely used instrument, however, is the Geiger-Müller counter, which is generally associated with auxiliary circuits for recording, including scaling and coincidence circuits. The nature of fluctuations in counts is important to realize. Rough guides for estimating errors due to them are given. The cloud chamber and photographic recording are two methods of importance, the former in nuclear research, the latter in an increasing range of application to applied problems. Scintillation counters find use for fast counting and for gamma-ray counting at high efficiencies.



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## 4. Methods of Accelerating Atomic Particles

This chapter will deal with means for accelerating charged particles to high energies. Such apparatus can be readily classified into two groups: (1) direct or "single-transit" accelerators and (2) additive increment or "multi-transit" accelerators. In the first type of apparatus the final particle energy is the product of the charge on the particle times the voltage developed by the machine. Due to insulation problems 12 million volts represents the maximum that has so far been attempted with an arrangement of this kind. The additive increment accelerator on the other hand develops relatively small voltage itself. Yet by treating the charged particle to this voltage kick many times in rapid succession tremendous final energies are feasible. Particle energies of 400 Mev have been attained by this method, and projects now under way have as their goal particle energies in the multibillion volt region.

### DIRECT ACCELERATORS

#### Electrostatic accelerators

The simplest and one of the earliest methods developed for producing the high voltages necessary for transmutation work is a streamlined version of the age-old electrostatic machine. Inasmuch as no ion source giving more than a few hundred microamperes of ions continuously has been perfected, the power actually utilized in the production of the most intense available beams is relatively small. This means that, although the accelerating device must develop a high voltage, no large current capacity is necessary. These requirements are admirably suited to the capabilities of an electrostatic generator. Van de Graaff was the first to recognize the possibilities of such a machine, and the present-day electrostatic apparatus as applied to atomic pro-



jectile work is a direct outgrowth of his development research and bears his name.

A description of the largest machine of this type ever constructed will illustrate the principles of operation. This apparatus was originally assembled at Round Hill, Massachusetts, where an airship

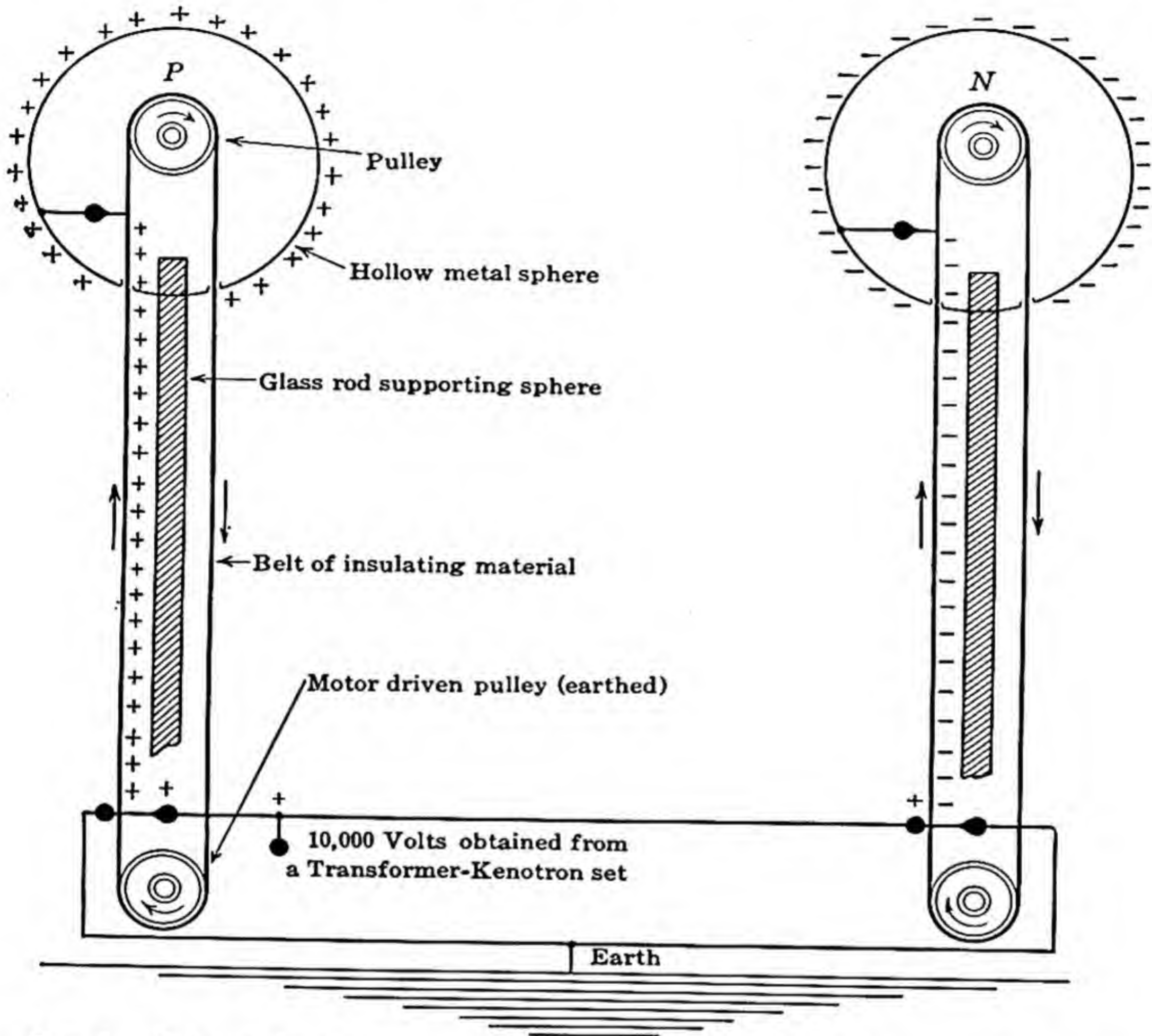


FIG. 1. Schematic diagram of the Round Hill high-voltage installation.

hangar served as its home. Later a special building was constructed on the campus at the Massachusetts Institute of Technology, where it was reassembled in a modified form. In the Round Hill installation two large metal spheres, each 15 feet in diameter, were mounted 40 feet in the air on insulating pillars. It is pictured diagrammatically in Fig. 1. A wide endless belt of silk or paper, driven by motors and pulleys, runs from the floor up inside each sphere. A transformer-rectifier set produces a voltage of 10 kilovolts. Negative electricity is sprayed on one of the moving belts from a sharp-pointed metal



comb, and positive charges are similarly put on the other belt. These belts then carry the charges upwards and within the spheres, where a similar metal comb removes the charge from each belt. The comb is connected to the sphere itself, and the collected charge is transported to the outside of the sphere. This must be the case since an elementary theorem in electrostatics states that no free charge ever remains on the inside of a hollow conductor. As a result of this, no charge ever accumulates on the comb, and hence, regardless of how much charge rests on the exterior, the charge sprayed on the belt below will always be taken off above. Thus,

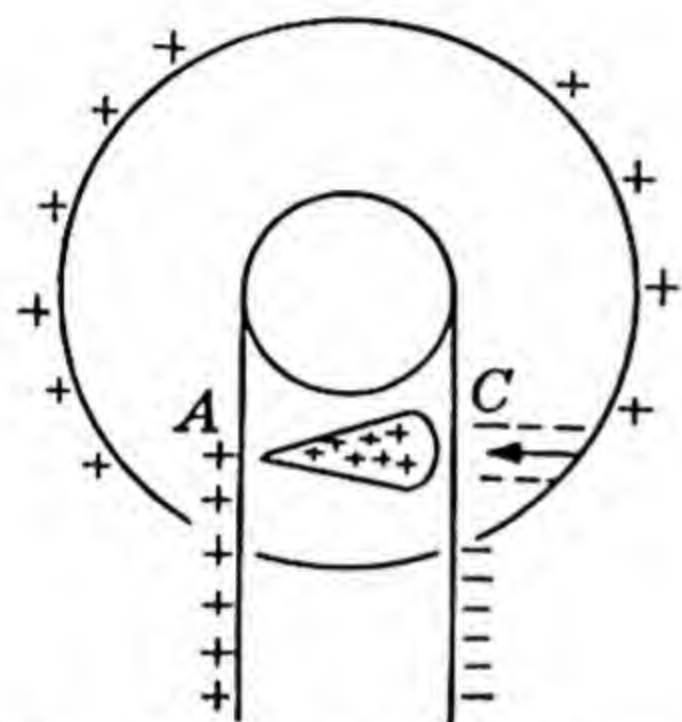


FIG. 2. Method of increasing belt efficiency by carrying negative charge down as well as positive up.

as more electricity is sent up the belt, the potential of the sphere rises until the charge lost to the air through corona discharge equals that coming up the belt. To make this equilibrium voltage as high as possible it is desirable to have wide belts and run them as fast as is practicable.

Another trick is employed which doubles the amount of charge separated in any one revolution of the belt. This is achieved by having the descending belt carry an equal quantity of charge opposite in sign to that which the ascending portion carries. It is accomplished in the following manner, as illustrated in Fig. 2. The rising belt carries a positive charge which is sprayed on below. On reaching A the sharp-pointed collector acquires a charge from the belt much as a vacuum cleaner sucks up dirt from a carpet. As this insulated collector assumes a higher and higher positive potential, negative electricity is attracted to the pointed rod C which is connected to the sphere. The resulting corona sprays negative charges on the descending belt. Thus each revolution of the belt is twice as efficient in charging the sphere as when the conventional scheme is used.

With this huge apparatus it was found possible to charge one sphere positively to well in excess of 2.5 million volts, and the other to a roughly equal negative potential. Now, by connecting an evacuated tube between the two spheres, ions produced at the positively charged sphere could be accelerated down the tube to an energy in excess of 5 Mev. These ions are produced in a discharge tube, into which gas, composed of the proper atoms, is continually admitted at a low pressure. A small hole drilled in the discharge tube allows the ions to



emerge into the accelerating tube, where they are whisked down the tube towards the target. If we prefer to accelerate electrons all that is necessary is to interchange ion source and target, the former being replaced by a simple electron gun. Figure 3 shows the accelerating tube in place. A short description of this tube is quite in order, since it is a common feature of all direct accelerators.

The tubular "doughnuts" are connected to metallic cylinders placed inside the accelerating tube, and through these cylinders the ion beam

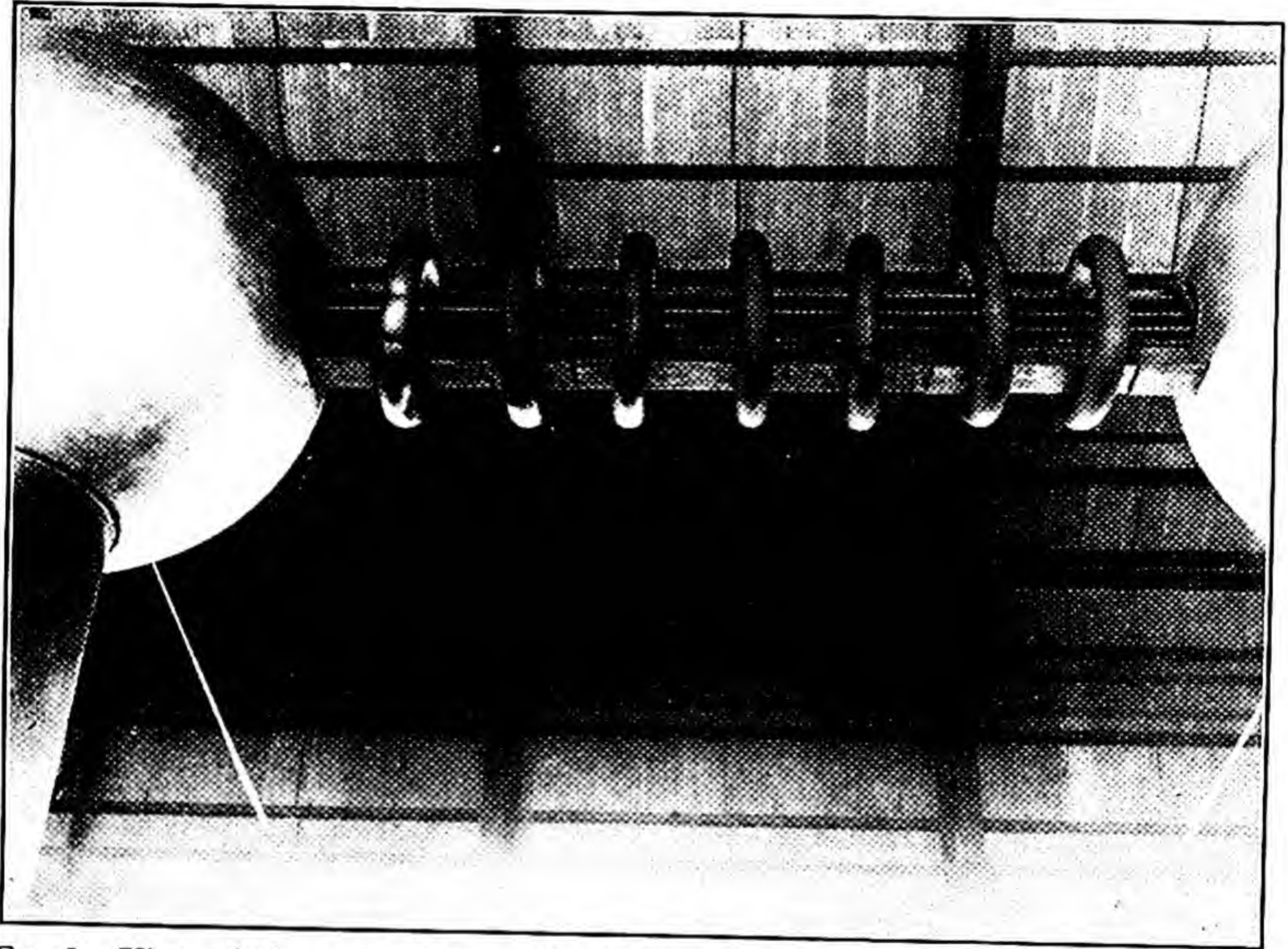


FIG. 3. View of the accelerating tube in place between the spheres of the Round Hill generator. (Photograph by Dr. L. C. Van Atta.)

moves. The doughnuts carry a corona discharge from one sphere to the other, thus making for a uniform voltage gradient along the tube. The cylinders are separated by gaps and act as lenses to focus the ions toward the target. Figure 4 provides a close-up of a section of the tube.

In actual practice there is a potential difference of several hundred kilovolts between each pair of inner cylinders to which the doughnuts are connected. The electrical lines of force between adjacent cylinders follow the pattern indicated. Ions moving through these electrodes



are constrained to move along the lines of force and as a consequence are confined mainly to a path near the center of the tube. The reason for this is obvious. In moving across the gap from the interior of one cylinder to the next the particle gains an increment of energy proportional to the voltage difference between the cylinders. This gain in energy results in a corresponding gain in velocity ( $K.E. = \frac{1}{2}mv^2$ ), which means that the velocity of the particle entering the second cylinder is much greater than the velocity it had when emerging from the first. Now, the faster a particle is moving in an electric field, the smaller the effect which the field can have in deflecting it. One might

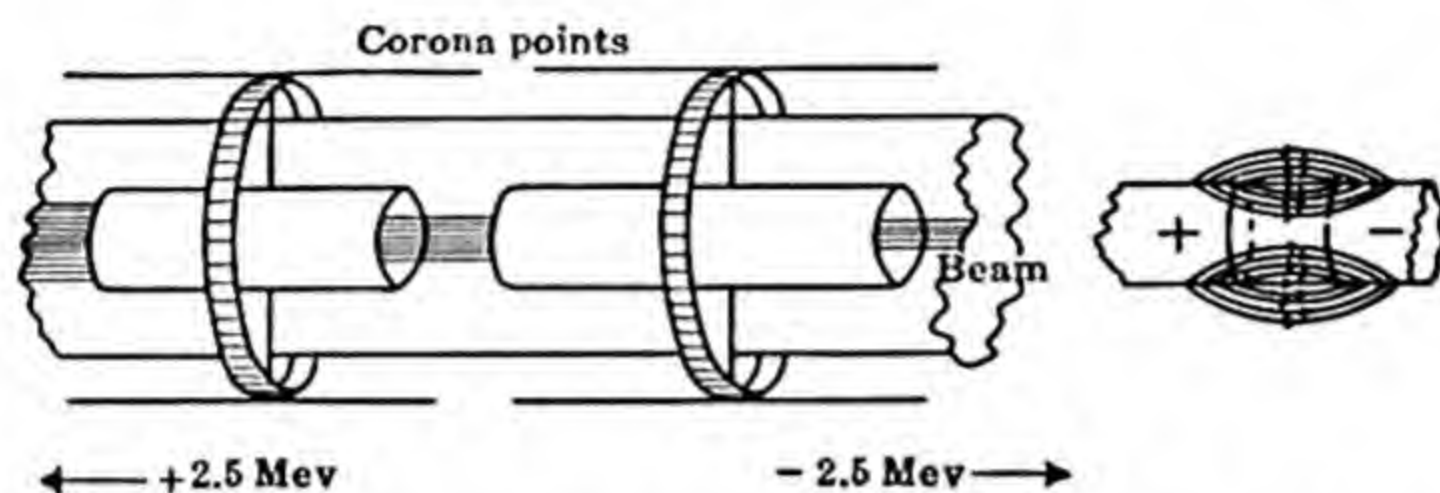


FIG. 4. Action of corona rings and points in distributing potential and focusing the beam. The small diagram shows the lines of force between the two sections. This field acts as an electrostatic lens.

visualize this in a non-technical way by arguing that a "slow" ion stays at any one place longer than a "fast" one and thus gives the field a longer time in which to act. Hence it is evident from Fig. 4 that the focusing effect exerted on an ion moving off center during the first half of its path across the gap will be greater than the defocusing effect experienced by the now faster ion during the second half of its journey between electrodes. This focusing action is so effective that, after careful adjustments have been made, the ion beam may have a cross section no larger than a dime. This facilitates the preparation of concentrated samples of radioactive materials.

In the modified installation at the Massachusetts Institute of Technology the two spheres are joined and charged as a single unit. A cross section of the new arrangement is shown in Fig. 5. A modified form of Van de Graaff accelerator was pioneered by Tuve, Hafstad, and Dahl at the Terrestrial Magnetism Laboratory of the Carnegie Institution at Washington, D. C. This apparatus proved very successful for voltage up to 1.2 million.

If a diatomic gas such as hydrogen is introduced into the discharge tube and if it were possible to view the resulting ion beam under a super-powerful microscope, we should notice that the beam is com-



posed of two types of projectile, one component comprising single particles and the other a number of dumbbell-shaped double units. The single particles would be protons, and the pairs would consist of molecular hydrogen ions. The molecular ion is in reality two protons

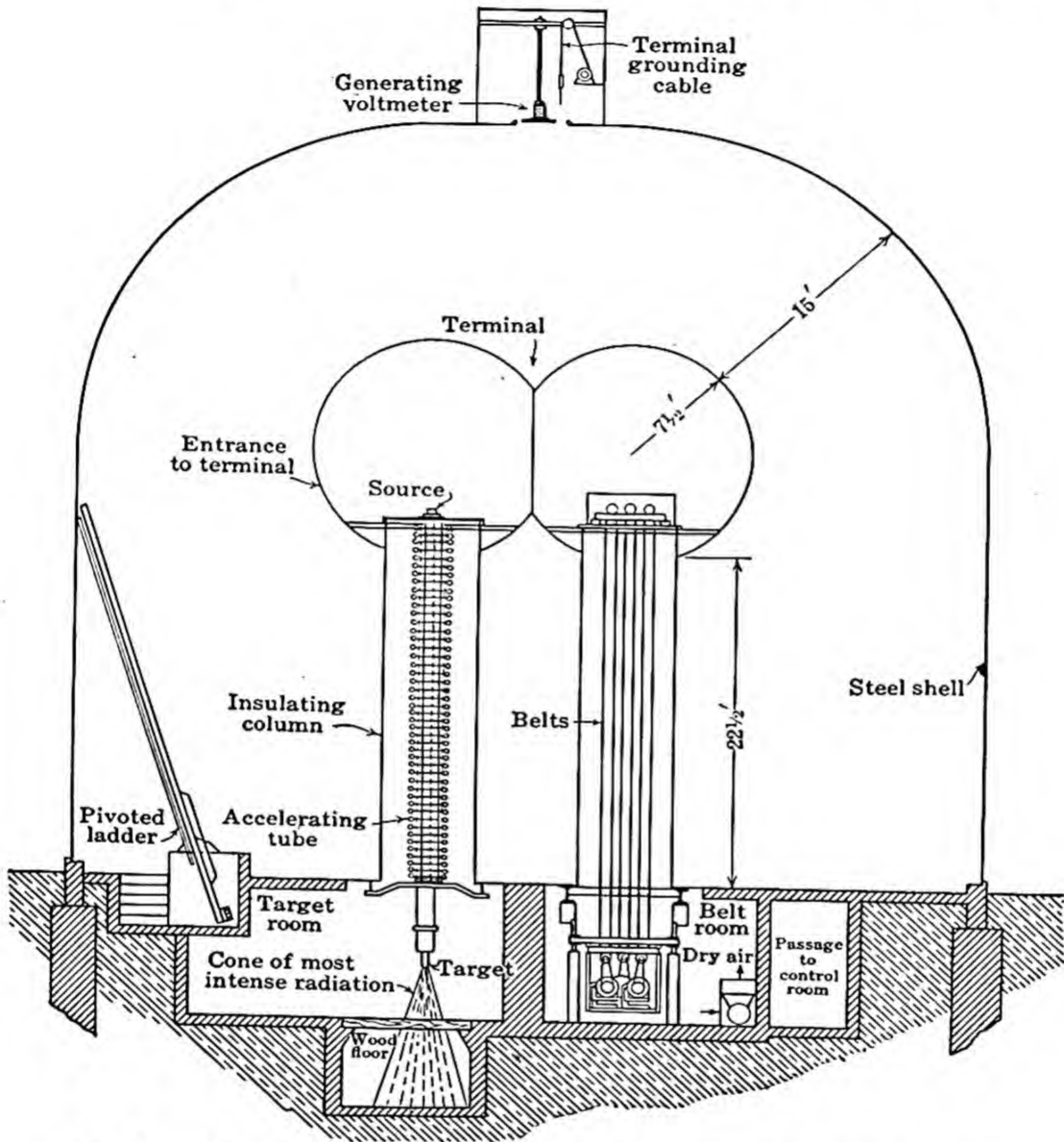


FIG. 5. Diagram of the Cambridge, Massachusetts, installation.

which share an electron. Since both types of projectile forming the beam possess effectively a unit positive charge, the kinetic energy given to each after passing down the accelerating tube will be the same. This requires that the molecular ions move with approximately seven-tenths the velocity of the protons. The impact upon striking the target splits each doublet ion into two protons. These "Johnny come



lately" protons possess only half the energy of those which traveled the length of the tube as such. As a consequence of this, two different energies are effective in producing transmutations when a target is exposed to the beam. For many experiments it is essential that monokinetic particles strike the target. This can be accomplished by superimposing a magnetic field at right angles to the path of the beam just ahead of the target. In such a field the molecular ions will be deflected somewhat less than the protons, and the initial beam will be separated into two parts, one comprising molecular ions, the other protons. The target can thus be placed so as to intercept whichever fraction is desired.

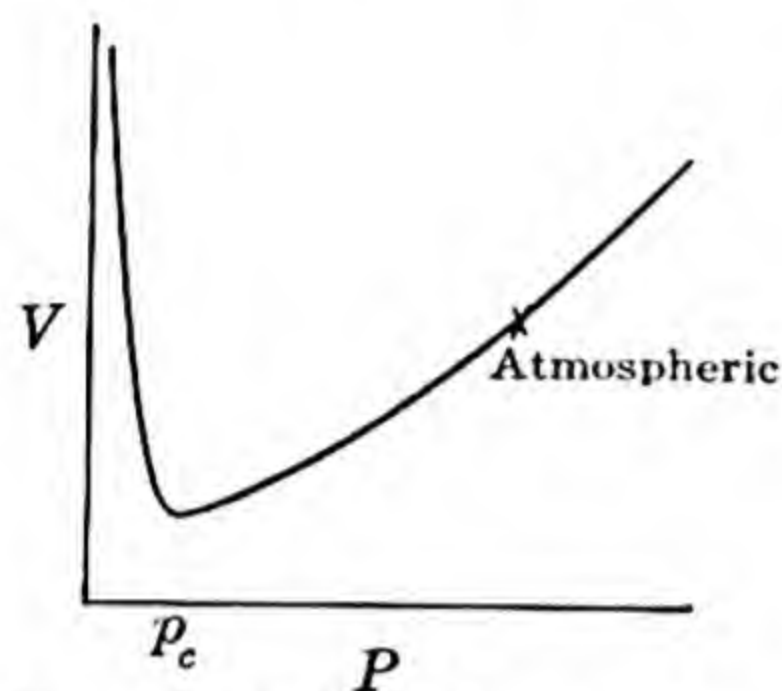


FIG. 6. Graph showing how the breakdown potential between two objects in air varies with the pressure. Provided that a critical pressure is exceeded, the breakdown voltage increases rapidly as the pressure is increased.

### Pressure electrostatic accelerators

The limiting voltage which can be attained by the sphere of an electrostatic-type apparatus is measured by the breakdown potential difference between it and surrounding objects at ground potential. A graph showing how the breakdown voltage between two objects varies with the air pressure is given in Fig. 6. It is clear from the curve that one can increase the ultimate voltage obtainable in either of two possible ways: (1) by evacuating the surrounding space to a point well below the critical pressure, or (2) by increasing

the pressure to several atmospheres.

The latter possibility has proved to be the more practical, and several installations which operate under pressures up to 10 atmospheres are now completed or in the process of completion. Foremost among workers in this phase of high-voltage machines has been Herb at the University of Wisconsin. A sectional view of his apparatus is shown in Fig. 7. The entire unit is enclosed in a steel tank built to withstand a pressure of 100 pounds per square inch. The sphere which usually serves as a high-potential electrode is replaced by the metal cylinder *E* supported by the Textolite cylinder *T*. The hoops *H* are of aluminum tubing spaced  $\frac{9}{16}$  inch apart. These hoops permit the flow of a small leakage current from the high-voltage electrode *E*, thus ensuring a uniform potential gradient from *E* to ground and discouraging sparking to the wall of the tank. The electrode is charged by the



system of belts illustrated. The accelerating tube is of conventional design. At atmospheric pressure the highest working voltage with this apparatus is about 500 kilovolts. However, at 8 atmospheres of air pressure the usable voltage is 2.1 Mev. Later investigations have shown that even larger voltages (2.4 Mev) are obtainable when  $\text{CCl}_4$  or  $\text{CCl}_2\text{F}_2$  vapor is introduced into the tank, and by various other improvements Herb, Turner, Hudson, and Warren have been able to push the voltage as high as 4 million.  $\text{SF}_6$  has been used at M.I.T. by Van de Graaff's group, and voltages up to 5.6 Mev have been obtained from a very compact apparatus.

The close control which one can exert on the voltage makes the electrostatic accelerator ideal for accurate nuclear work. McKibben

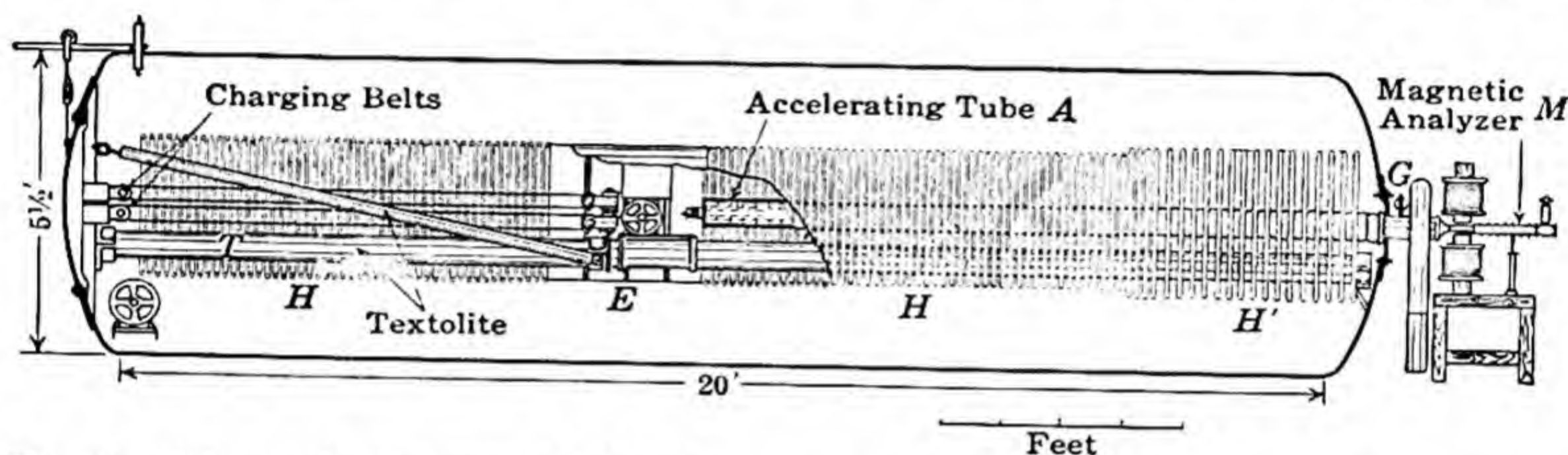


FIG. 7. Schematic diagram of Herb's pressure electrostatic generator. The hoops  $H$  ensure a uniform potential distribution along the length of the accelerating tube on the one hand and the charging belts on the other. This equipment is cheap and compact and can develop about 2.5 million volts.

has reported a scheme for maintaining the voltage of a 2.5-Mev accelerator constant to within  $\pm 1.5$  Kev, which is better than 1 part in 1600. This is done by making the analyzed beam control a variable corona leakage current to the high potential electrode and so automatically regulate the voltage.

The final development of this form of acceleration has not yet been reached; machines designed for 12 Mev are now under construction at Los Alamos and M.I.T. The High Voltage Engineering Corporation of Cambridge, Massachusetts, now offers for sale a commercial model of the Van de Graaff apparatus.

### Voltage multiplication methods

We now turn our attention to the scheme used by Cockcroft and Walton in England for obtaining high voltages. With their machine they were the first to produce a transmutation with artificially accelerated particles. Their method depends on the process known as



voltage multiplication originally worked out by Greinacher. The same idea is utilized in many home radio sets. For example in Fig. 8

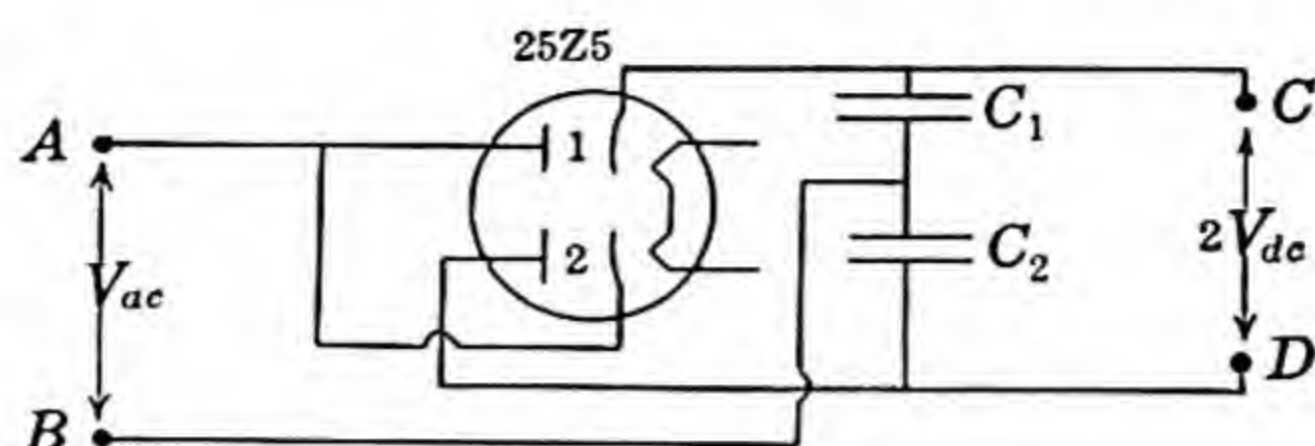


FIG. 8. Circuit showing how the Greinacher idea is applied in transformerless radio sets. The output voltage under no load is twice the peak input voltage.

when  $A$  is at its positive voltage peak, diode 1 becomes conducting, charging  $C_1$  to a potential difference  $V$ . A half-cycle later diode 2 conducts, raising the potential difference across  $C_2$  to  $V$ , but in the opposite sense to  $C_1$ . Hence there appears across  $CD$  a constant volt-

age  $2V$ . If a load is connected across  $CD$  this voltage decreases somewhat.

Elaborating on this idea Cockcroft and Walton used the circuit shown in Fig. 9 to get a final voltage six times that supplied by the transformer. The accelerating tube is connected across  $AB$ . At high voltages there is always a certain amount of corona loss, and of course a slight amount of power is needed for accelerating the ions. The actual voltage multiplication in their apparatus turned out to vary between 5 and 6.

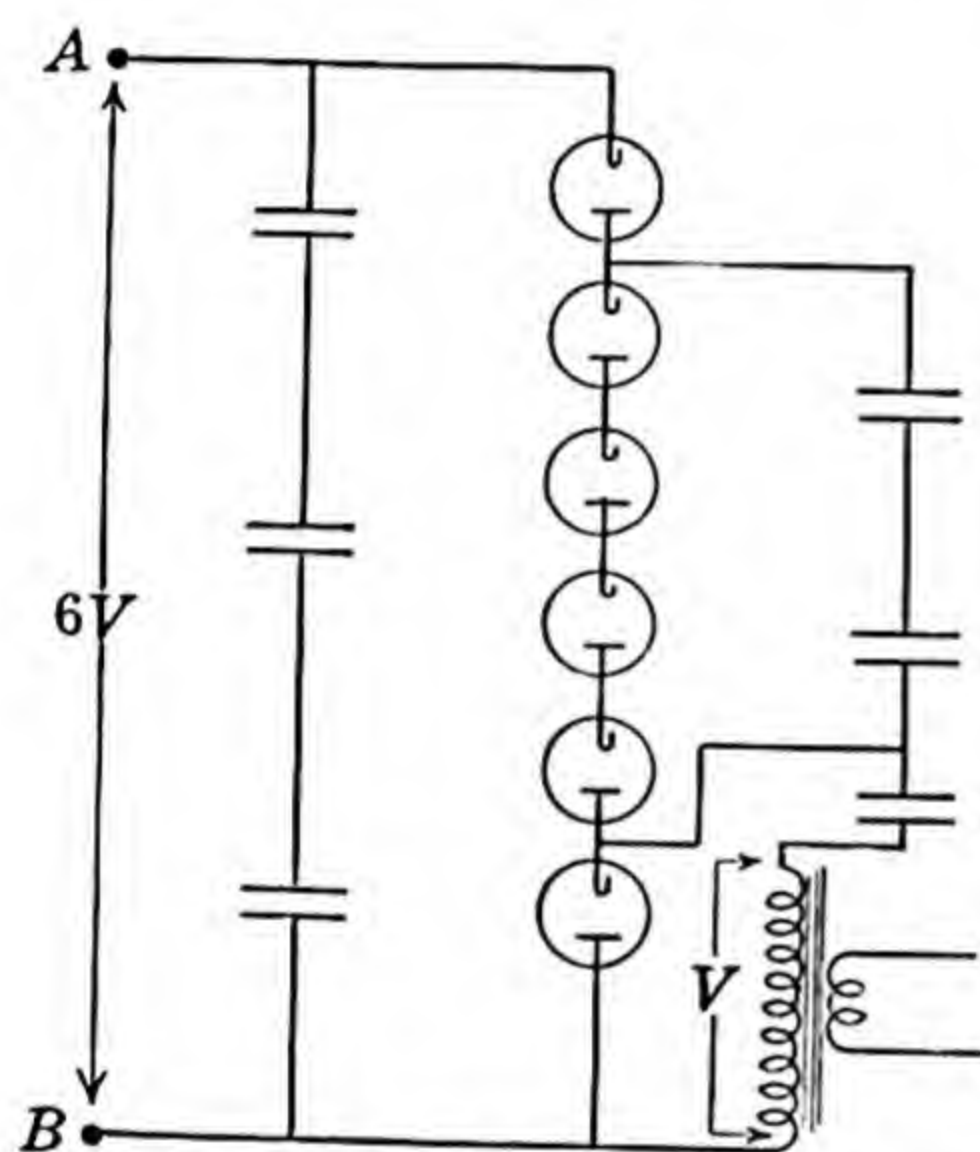


FIG. 9. Circuit used by Cockcroft and Walton which yields a voltage between five and six times that supplied by the transformer. The accelerating tube is connected across  $AB$ . This is an extension of their original installation which only quadrupled the transformer voltage.

Four transformers are linked in cascade as indicated, a portion of each secondary exciting the primary of the following stage. Thus the secondary of each transformer operates at a root mean square

### Cascade transformer methods

Early in the 1930's Lauritsen and his associates modified a high-voltage x-ray installation at the California Institute of Technology to serve as an ion accelerator. The essentials of this machine are shown in Fig. 10.



(rms) voltage of 250,000 above that of the preceding stage. This means that the fourth transformer will be raised to a potential 1 million volts above ground. The one major disadvantage with this arrangement is that the voltage is not constant but alternates between 1 million volts positive and negative. Consequently, positive ions will be accelerated only during the positive half-cycle, and not all of them will have the full million electron volts of energy. This means a lower efficiency for transmutations than would result from an equal number of ions accelerated in a 1-million-volt constant-voltage apparatus. In addition to this, any electrons present in the free state will be accelerated throughout the negative part of the

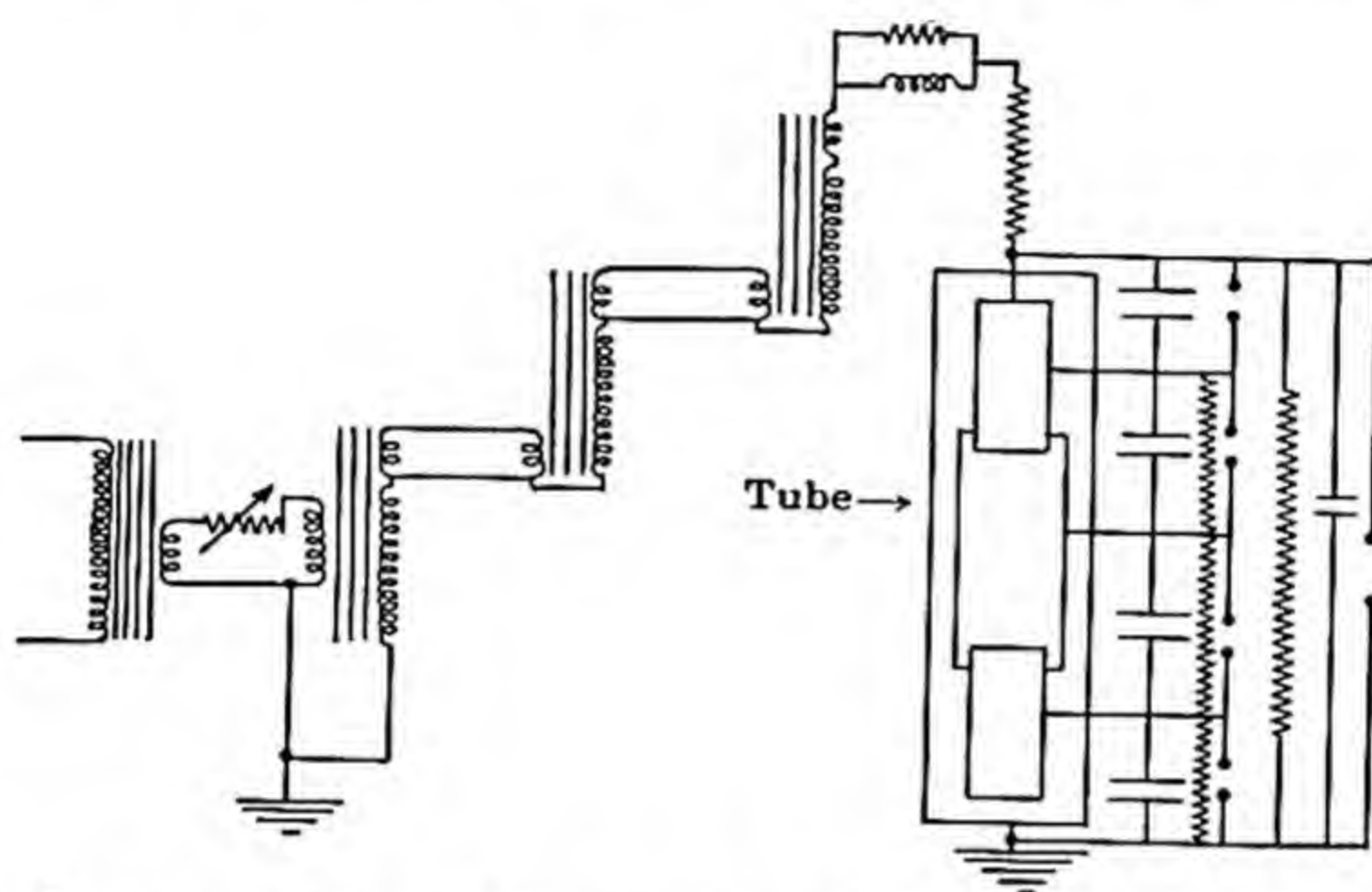


FIG. 10. Transformers in cascade. Part of the secondary of each transformer operates the primary of the next, until high voltages are built up. The tube is in three sections with the voltage applied from a divider as shown.

cycle, and these on striking the target will give rise to penetrating x-rays which can prove quite troublesome. Nevertheless, many valuable experiments have been carried out with this apparatus, and it seems especially suited to work involving a Wilson cloud chamber.

## MULTI-TRANSIT ACCELERATORS

### Linear multiple accelerator

The simplest apparatus utilizing the indirect method is the linear multiple accelerator, first demonstrated by Wideroe and developed in America by Sloan and Lawrence. The name aptly describes its mode of operation. The main features are shown schematically in Fig. 11. By means of a fixed potential  $E_0$ , ions formed at  $S$  are drawn into the



first of several coaxial cylinders which are enclosed in an evacuated chamber. Alternate cylinders are connected to the ends of a coil which is inductively coupled to the tank circuit of a high-frequency oscillator. An alternating potential of several thousand volts is thus produced across the gap between the cylinders. The ions present in the gap during a favorable half-cycle will be accelerated across the gap. Let us assume that the cylinder length and spacing are chosen so that a particle which receives a voltage kick  $V_e$  will traverse the

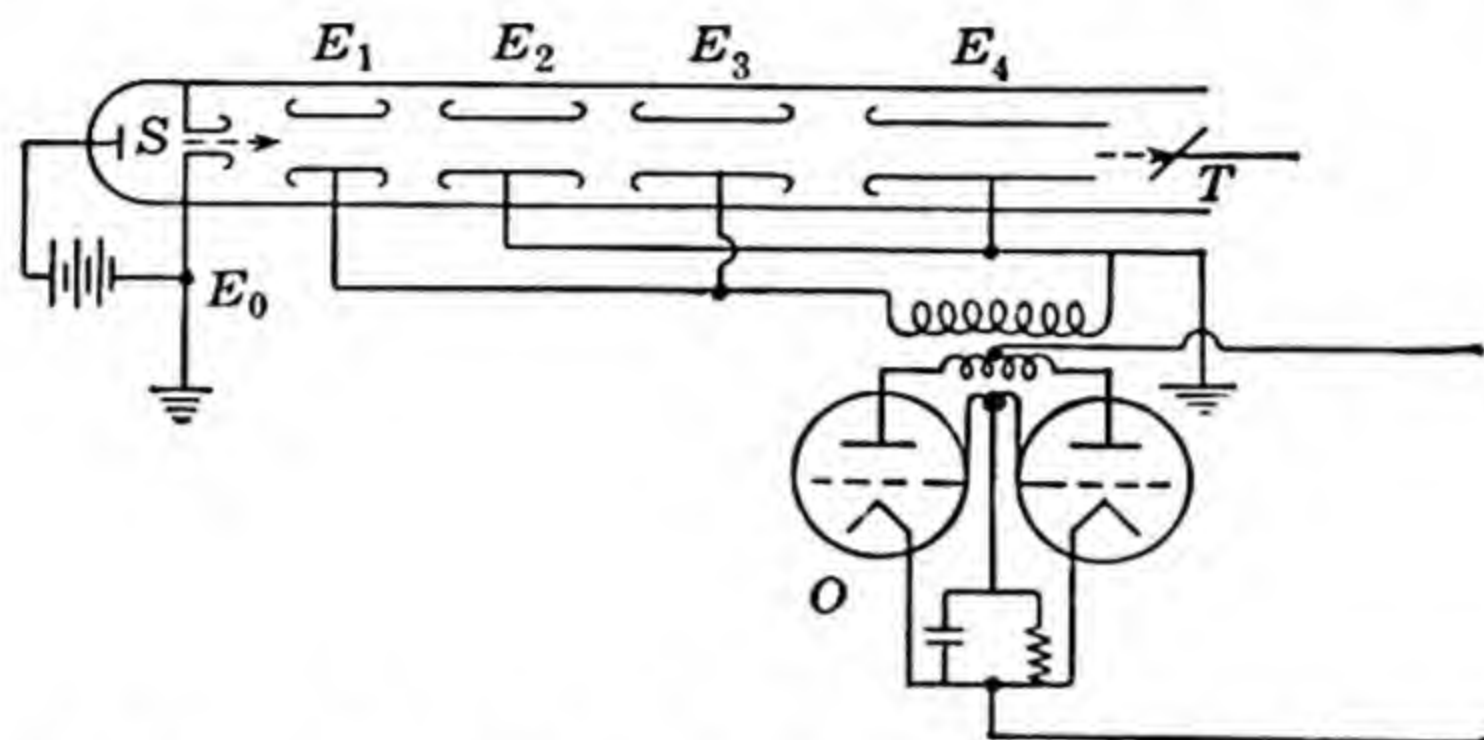


FIG. 11. Linear multiple accelerator. In this apparatus a radio-frequency voltage is impressed on alternate cylinders, whose lengths are carefully adjusted. An ion which is accelerated from  $E_0$  to the first cylinder is also accelerated in the next gap, since the time of transit of the first cylinder is just that of a half-cycle. The equipment becomes too long and requires too much power for the development of voltages in excess of a million.

first cylinder in a time equal to a half-period of the oscillator. This means it will enter the second gap when the voltage between cylinders is again  $V_e$  and will receive an additional energy increment of this magnitude. As the particle gains in velocity it is obviously necessary that successive gaps be spaced a correspondingly greater distance apart. Originally one great disadvantage of this apparatus was that with the radio frequencies available in 1932 unreasonably long tubes would have been needed to produce light ions with energies in excess of 1 Mev. By using 36 accelerating cylinders Sloan and Coates were able to obtain singly charged mercury ions of almost 3 Mev, although the ion currents were very small. Such heavy ions are not efficient in causing transmutations since in colliding with a nucleus most of the energy is spent setting the target nucleus in motion and thus only a small fraction is available for effecting a transmutation.

Another glaring defect in this instrument is the difficulty of trying to achieve both "axial" and "phase" stability. This can be appreciated by reference to Fig. 12. It is clear that particles reaching the



gap at both  $t_1$  and  $t_2$  will receive the proper push  $V_e$ . However, the  $t_1$  particles cross the gap while the electric field is increasing. Hence in traversing the gap they will receive a net defocusing effect (more defocusing over the second half of the gap than focusing over the first half), and any off-center motion they possess will be accentuated. In other words, they lack axial stability. The  $t_2$  particles have axial stability since they move across the gap while the field is decreasing, but they in turn lack *phase stability*. To understand this latter term let us concentrate on a  $t_2$  particle. It should enter the next

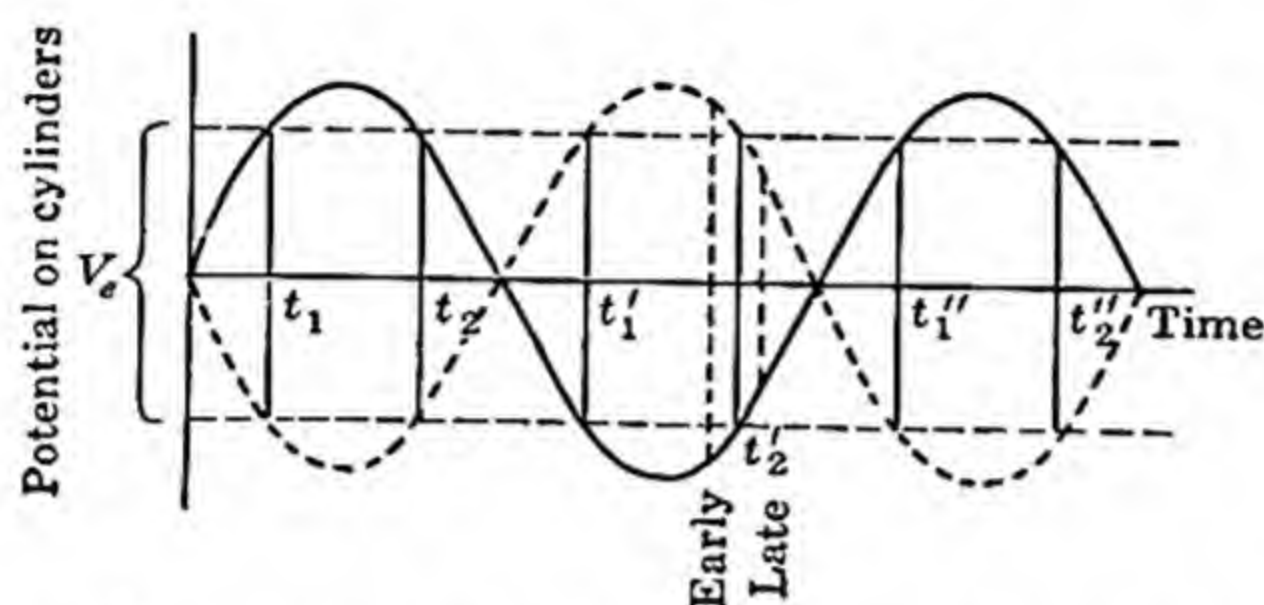


FIG. 12. Solid line shows potential on odd-numbered cylinders; dashed line, potential on even-numbered electrodes; both as a function of time. Vertical distance between curves is potential difference at time considered. Particles entering gap at  $t_1$  and  $t_2$  will receive proper acceleration. However,  $t_1$  particles lack axial stability, whereas  $t_2$  particles lack phase stability.

gap at time  $t_2'$ . Should it be delayed slightly it will arrive at a time later than  $t_2'$  and get a smaller kick than it should, thus throwing it even further behind at the following gap. If it should be ahead of schedule it will get too large a boost and hence be even earlier at the succeeding gap. A similar analysis applied to  $t_1$  shows that, conversely, any deviation in phase will effectively correct itself so that the  $t_1$  particles will tend to keep in step with the field.

With the invention of the cyclotron, interest in linear multiple accelerators disappeared until the radar techniques of World War II made it appear feasible to obtain high-energy protons in a reasonable space. In 1948 Alvarez completed an installation at the University of California which produces 32-Mev protons in an apparatus 40 feet long. The accelerator is pictured in Fig. 13a, and although it scarcely resembles the apparatus sketched in Fig. 11 the basic principle is still the same. Proton pulses of 300-microsecond duration are given a preliminary acceleration to 4 Mev in a pressure Van de Graaff machine and are then fed into the first stage of the linear accelerator. Those protons entering at the proper stage of the radio-frequency



cycle are compressed into a tight bunch and accelerated down the tube. The 40-foot cylindrical cavity is energized by 27 radio-frequency oscillators operating at 202.5 megacycles, each capable of delivering

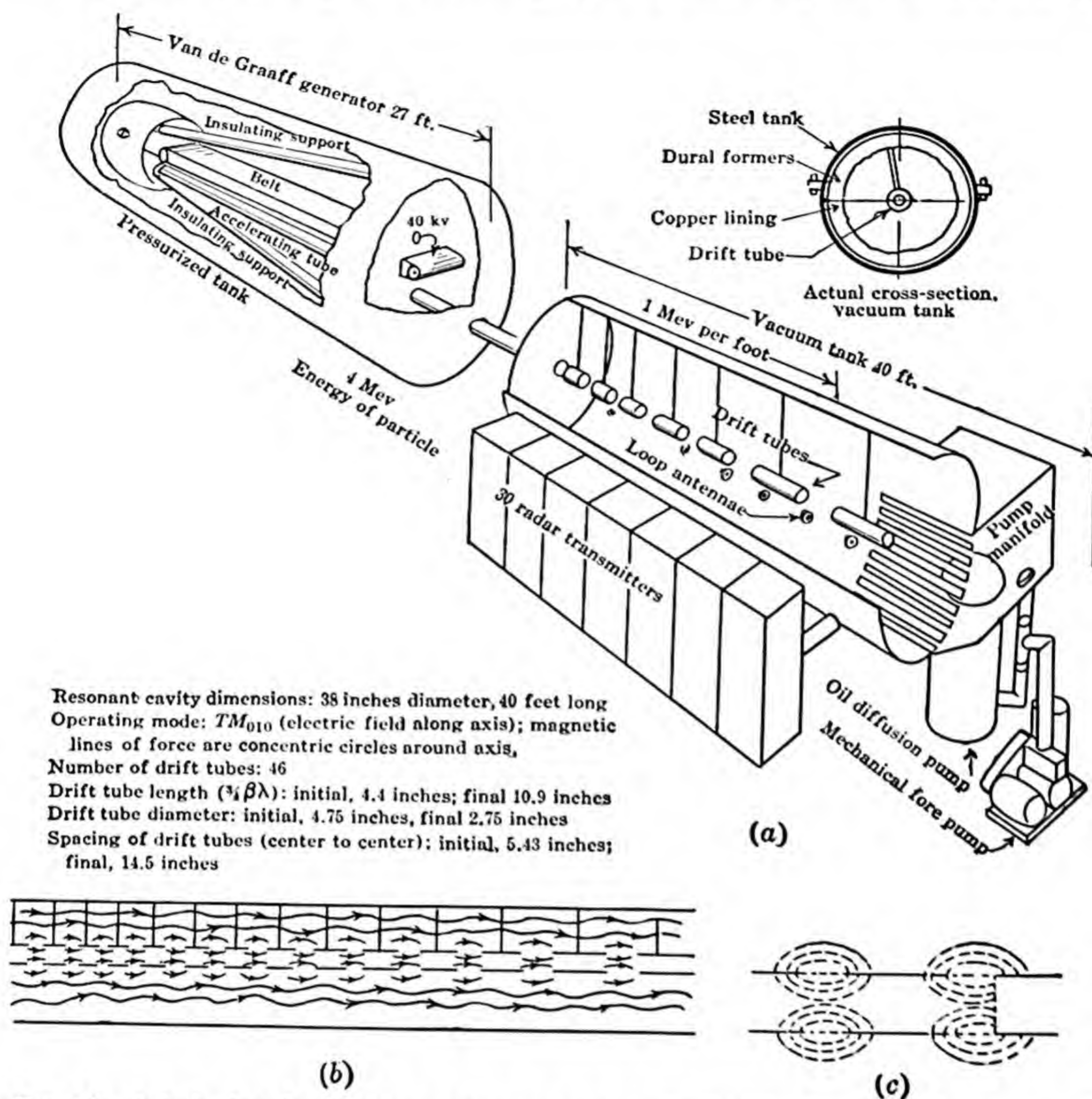


FIG. 13. (a) A sketch of Alvarez' linear accelerator which has yielded protons of 32 Mev (adapted from *Chemical and Engineering News*, Vol. 25, p. 227). (b) An instantaneous view of the electric lines of force in the vacuum tank. The magnetic lines of force are concentric circles about the tank axis as center. (c) The defocusing part of the electric field between drift tubes is eliminated through the use of a wire grid.

150 kilowatts of peak power in the form of 600-microsecond pulses at a repetition rate of 30 per second. No trouble was experienced in properly phasing the separate oscillators which lock into the resonant frequency of the cavity.



A standing wave field is established within the cavity, Fig. 13b showing an instantaneous view of the electric lines of force in a section. (A half-cycle later the arrows will be reversed.) Note that the field is in phase the full length of the cavity. A proton introduced into the first cavity at this instant will be accelerated. One radio-frequency cycle later it will be entering the next gap since the time required to traverse the distance between successive gaps  $\beta\lambda$  (time = distance/velocity =  $\beta\lambda/v = v\lambda/cv = \lambda/c$ ) is just one radio-frequency period ( $c = n\lambda$ ;  $n = c/\lambda$ ;  $T = 1/n = \lambda/c$ ). Thus it will receive a similar voltage kick at every gap. During the reverse phase of each cycle the protons coast along through the field-free interior of the so-called "drift tubes." The protons have a velocity of  $0.1c$  as they enter the first gap and are moving at a final speed of  $0.26c$  after negotiating the 40-foot cavity. Thus the drift tubes become longer and more widely separated down the cavity in order to satisfy the  $\beta\lambda$  requirement for gap spacing. Alvarez achieves phase stability by utilizing protons entering at  $t_1$  (Fig. 12) and overcomes the axial spreading by stretching thin wolfram wire grids across the entrance to each drift tube. This changes the field as shown in Fig. 13c and effectively prevents defocusing, although the grids do intercept a small portion of the proton beam.

The reader may wonder if it is possible to secure energetic electrons by this general method. The answer is yes, although the problems involved are somewhat different. For example, an accelerating electron quickly approaches the velocity of light, and phase stability is no longer operative. On the other hand this is not crucial since it is fairly easy to establish a standing wave pattern in coupled cavities with a phase velocity equal to  $c$  or load a wave guide so as to achieve the same phase velocity in a running wave. Neither is the absence of axial stability too important here inasmuch as the Lorentz contraction serves to make a long tube appear quite short to a relativistic electron, the apparent length being only  $\sqrt{1 - \beta^2}$  of the actual length.

Three separate schemes as sketched in Fig. 14 are currently being studied for the linear acceleration of electrons. The first being developed at M.I.T. by Slater involves a large number of resonant cavities mutually coupled and powered by an array of synchronized magnetrons. A standing wave pattern is set up within the cavities with the electric vector along the axis ( $TM$  mode). This pattern can be viewed as the superposition of two waves traveling in opposite directions through the cavity array, each with phase velocity  $c$ . The electrons ride the crest of the wave, moving from left to right, while the



reverse wave, contrary to what might be expected, produces only unimportant perturbations. Just as a stretched cord can vibrate in numerous different ways, so can a series of resonant cavities be excited in many different modes. The big problem here is to match the cavities so that the desired mode alone will be excited and all others

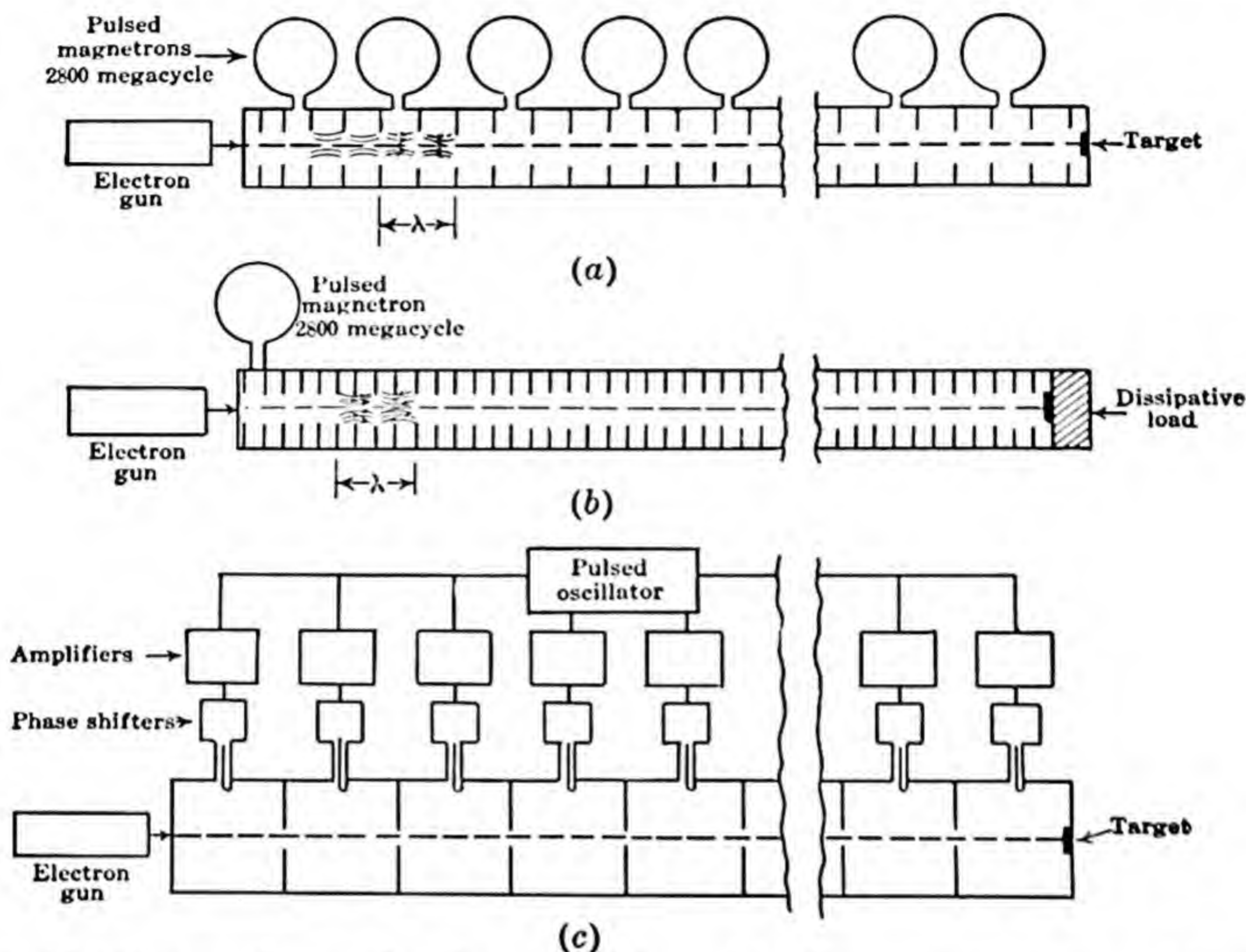


FIG. 14. Three schemes for the production of high-energy electrons. The first may be viewed as a series of tightly coupled cavities or as a wave guide in which a standing wave is established. The second method involves a running wave through a loaded guide which reduces the phase velocity to  $c$ . The third scheme involves the ganging of uncoupled resonant cavities driven by amplifiers excited by a common oscillator. Proper phasing is accomplished by means of phase shifters.

suppressed. In operation 50- to 100-Kev electrons are introduced into the first cavity where the transition to relativistic speeds leads to an effective bunching. The electron bunch then moves down the tube with velocity  $c$ , gaining in mass and consequently in energy. The word ponderator has been suggested as a more suitable name for this device than the term accelerator.

Figure 14b depicts the traveling wave scheme being used successfully at Stanford and Harwell (Great Britain) to produce energetic electrons. The long cylindrical wave guide is loaded with circular



irises to reduce the phase velocity to  $c$ . Microwave energy is introduced at one end and absorbed at the other, so no energy is reflected. It is believed that a length of 50 feet can be fed efficiently in this way. Twenty-Mev electrons should be obtainable over this length when the wave guide is pulsed by a single HK-7T magnetron (750,000-kilowatt peak power). Figure 15 shows a 2-meter experimental apparatus built by Fry at Harwell which has produced 15 microamperes of 4.2-Mev electrons. Hansen at Stanford was able to obtain 4.5-Mev electrons in a similar apparatus 9 feet long.

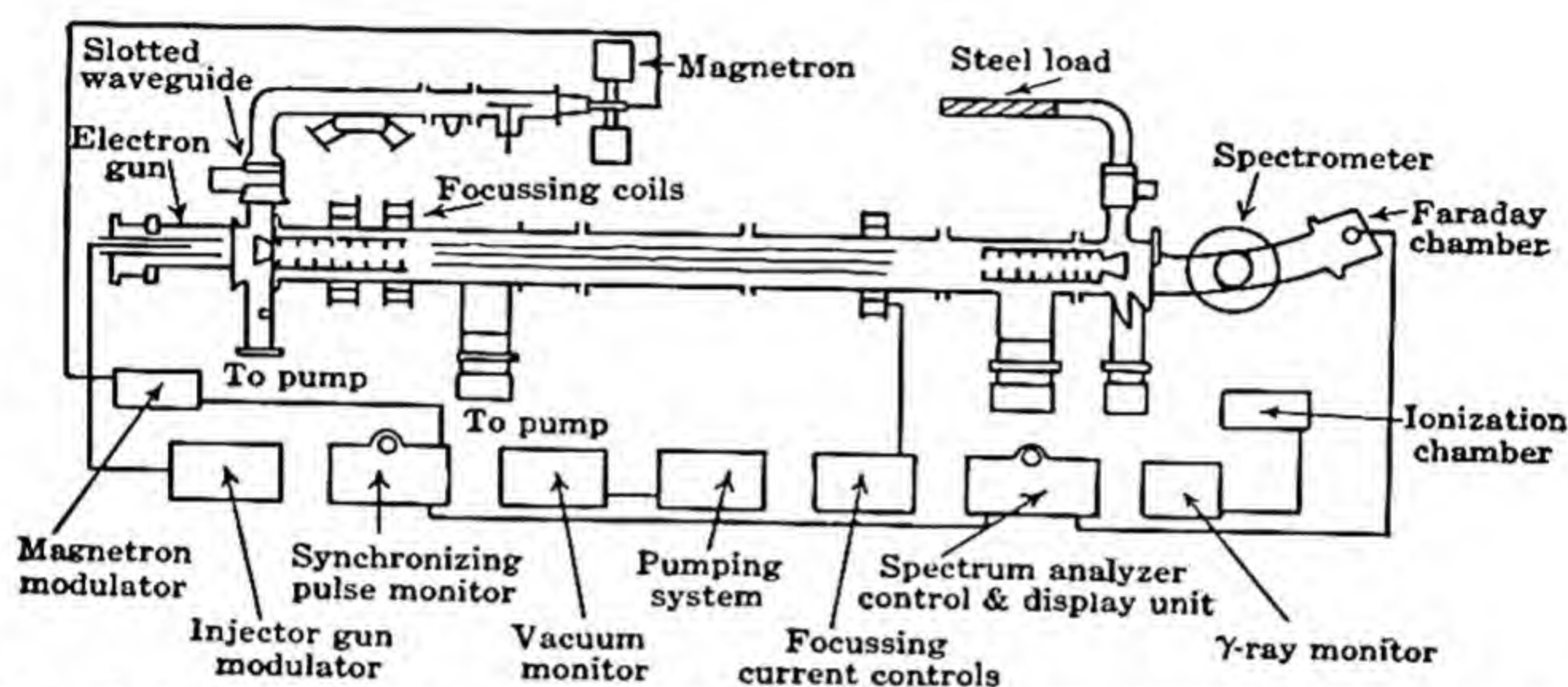


FIG. 15. Detailed sketch of a traveling wave accelerator which produces 15 microamperes of 4.2-Mev electrons. Constructed by Fry et al. at Harwell, Great Britain.

Figure 14c illustrates the method employed by Schultz at Yale to accomplish the same end. He utilizes a large number of resonant cavities in line though mutually uncoupled. A master oscillator drives an array of power amplifiers (X126P) at 600-kilowatt peak power, each of the latter serving to excite a single cavity. The highest operating frequency for such an arrangement is 580 megacycles. A phase shifter between each amplifier and cavity permits the whole group to be properly phased. An energy increment of 1.2 Mev per stage is anticipated, and there is no theoretical limit as to the number of stages that can be ganged.

Regarding the future role to be played by linear devices, it appears that their greatest utility will be in the Bev (billion electron volt) electron range. For lower electron energies the betatron and synchrotron may prove more economical. The Van de Graaff machine, cyclotron, frequency-modulated cyclotron, and proton synchrotron appear to blanket the entire range of heavy-particle energies.



### The betatron

This device, the first successful apparatus for multiple acceleration of electrons, was announced by Kerst in 1940 though others had speculated earlier on its possibilities. It has now been developed sufficiently by Kerst at the University of Illinois to yield 315-Mev electrons. Two concerns, the General Electric Company and Allis-Chalmers, are producing commercial models. Though in age the betatron is years younger than the multiple linear accelerator, it is based on a principle as old as that of the transformer. In fact the transformer principle is essentially the basis of betatron operation. The idea can be explained as follows: Suppose that we have an iron cylinder which can be magnetized by current through a surrounding coil of wire. Assume a copper ring enclosing the cylinder, and imagine the field increasing. Everyone is familiar with the fact that an electromotive force is set up in the copper which causes a current to flow; such an induced current is quite commonplace. Now imagine the copper ring replaced by a frictionless tube into which an electron could be introduced while the field is increasing. This electron experiences a force; in fact, such a force is the basis of the electromotive force producing the current in the wire, and this force causes the electron to be accelerated. Now it will not matter how fast the electron is moving, or where it is; provided that it remains at the same radius from the cylinder, the force will persist. The force on the electron depends on the field strength at the orbit and the rate of change of the magnetic flux within the orbit.

Suppose now that the magnetic field has a uniform rate of change for  $\frac{1}{1000}$  second—a short interval of time. Suppose also, to make the matter definite, that the electric field strength exerted on the electron while the magnetic field is changing is 1 volt per centimeter. This is  $\frac{1}{300}$  electrostatic unit of electric field, so that the electron experiences a force of  $e/300$  dyne. This produces an acceleration of  $e/300m$  cm per second per second, which the reader can easily verify for himself is of the order of  $10^{15}$  cm per second per second. In  $\frac{1}{1000}$  second the electron is thus readily accelerated beyond the velocity of light, if its mass remains unaltered. However, as the velocity approaches that of light the mass of the electron increases, so that as a rough approximation we can suppose that the electron is quickly accelerated to very nearly the velocity of light and stays at that figure for most of the  $\frac{1}{1000}$  second. It will therefore travel approximately  $3 \times 10^7$  cm. Now an electron which falls this very considerable distance in a field of 1



volt per centimeter will acquire the very large kinetic energy corresponding to the charge times the field times the distance traveled, which is easily seen to be 30 Mev. If, then, this kind of trick can be worked, the rewards will be considerable; it remains to exert a little ingenuity.

If the reader is astute he will see that ingenuity is needed to solve two problems. The first is the obvious one of the frictionless tube; the second is the problem of focusing. Both these problems must be solved or the "induction accelerator" will be useless. The first is not so bad, because we know that an electron will move in a circular path in a magnetic field; all that we need is to find such a field that an electron will move in the *same* circular path while the magnetic field changes. Thus if we use electrostatic units for the charge on the electron it can be shown that the momentum of the electron is given by

$$mv = \frac{Hre}{c} \quad (1)$$

and if  $\phi$  is the magnetic flux enclosed by the circular path the tangential electric field is

$$E = \frac{\dot{\phi}}{2\pi rc} \quad (2)$$

Now in all these considerations we must remember that the mass of the electron is not constant; that  $mv$ , the momentum, involves both quantities as variables. If we remember this and write the momentum as  $p$ , then the second law of motion gives us

$$\dot{p} = \frac{e\dot{\phi}}{2\pi rc} \quad (3)$$

from which we deduce that, if in a certain time interval the flux enclosed changes from  $\phi_0$  to  $\phi$ , the momentum developed will be

$$p - p_0 = \frac{e(\phi - \phi_0)}{2\pi rc} \quad (4)$$

which combined with equation 1 gives us

$$H - H_0 = \frac{\phi - \phi_0}{2\pi r^2} \quad (5)$$

Now if we consider the flux due to a *uniform* magnetic field  $H$  spread over the area of the orbit and call it  $A$ , we can write equation 5 as

$$\phi - \phi_0 = 2(A - A_0) \quad (6)$$



which means that for a constant electron orbit of radius  $r$  the actual change in flux through the orbit must be twice that which would obtain if the magnetic field had its orbital value over the enclosed area. This is the well-known betatron 2:1 requirement, which can be realized by introducing an air gap or gaps in the central core.

The problem of focusing is more difficult. The electrons travel a total distance of several hundred miles, yet we propose to direct them at a target little more than 1 sq cm in area. Such a stringent requirement would make the operator of a Norden bombsight shudder in dismay. However, it turns out that there is a tendency for the electron to follow a path which executes a *damped* oscillation about this stable circular path, provided the magnetic field near the orbit diminishes with radius according to  $H \sim 1/r^n$ , where  $n$  lies between  $1/2$  and 1.

The focusing conditions were worked out by Kerst and Serber. Guided by these, Kerst completed the first induction accelerator in 1940, which produced electrons of 2.3 Mev. Larger machines quickly followed. Figure 16a illustrates the basic structure of a conventional betatron. The electrons are injected into the vacuum chamber from a "gun" of the type familiar in beam power tubes. Under the influence of the guide field the electrons turn into the equilibrium orbit  $r_0$ , where they continuously acquire energy as a result of the increasing flux within the orbit. One way of displacing the beam after acceleration involves a current pulse through auxiliary turns  $C$ , in such direction that the flux within the orbit is increased while the field at  $r_0$  remains unchanged. This causes the electrons to spiral outward, striking a metal target and generating energetic x-rays. If desired, the electron beam can be brought outside the accelerating chamber through a simple laminated iron channel, which provides a relatively field-free space. This permits the beam to be piped from the betatron to areas more accessible for experimental work.

In order to secure economically the varying magnetic field needed in a betatron, the exciting coils are made the inductive part of a resonant circuit; that is, banks of condensers are connected across the windings. The resonant circuit is coupled to a source of alternating current, the latter having to supply only enough power to compensate for  $I^2R$  and magnetic losses plus the minute amount needed to accelerate the electrons. In conventional betatron operation the electrons are injected at point 1 (curve accompanying Fig. 16a), at which time the fields both at and within the orbit are passing through zero. The electrons are accelerated over a quarter of the magnetic cycle, reaching maximum energy at 2.



A modification known as direct-current biasing can be employed to permit acceleration over nearly a half-cycle. This is done by superposing a direct current on the alternating current, the direct current

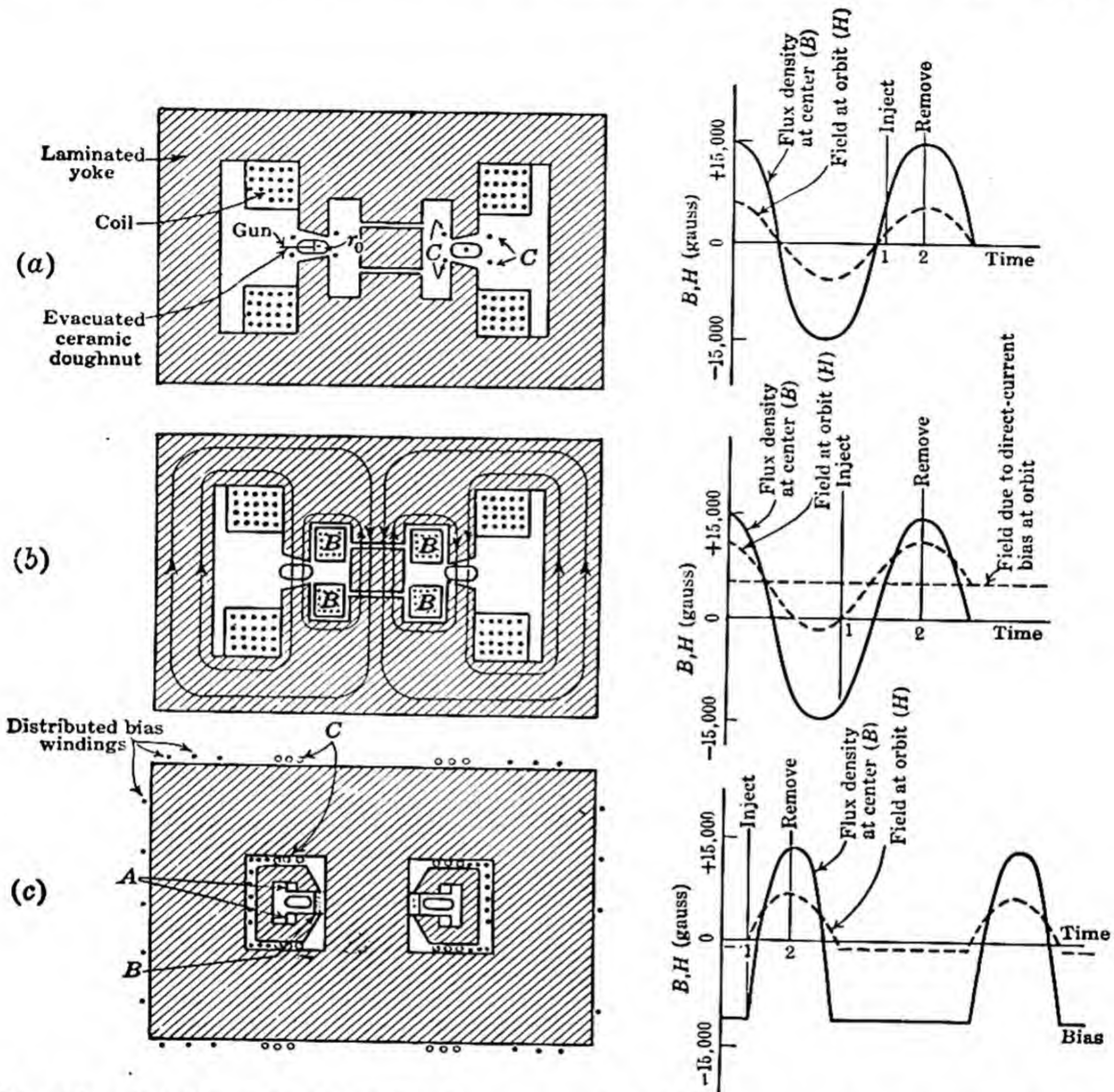


FIG. 16. (a) Simplified sketch of a conventional betatron. The magnet must be laminated to overcome the perturbing effects of eddy currents in the iron. The adjoining plot shows how the magnetic field at the orbit ( $H$ ) and flux density in the center ( $B$ ) vary with time. Acceleration is carried out over one-quarter of the alternating-current cycle. (b) Sketch of a betatron equipped with bucking coils. The direct current in the bucking coils produces a flux which cancels that due to the main coils within the orbit, but the two direct-current fields augment each other at the orbit. Here acceleration extends roughly over a half-cycle. (c) Cross-sectional view of a flux-biased betatron. The central core is direct-current biased almost to saturation in a negative sense. Unidirectional current pulses serve to energize this machine.



being somewhat less than the crest value of the alternating current. The final electron energy is unchanged since the total flux change from zero field to saturation is the same as above. However, the magnetic field swing is reduced almost by half, and since hysteresis in the iron accounts for most of the power loss in a betatron the total power consumption can be greatly reduced.

If it were possible to cancel the direct-current bias flux within the orbit while retaining the bias field at the orbit, one could conceivably introduce the electrons while the central flux is near its maximum negative value and remove the beam when the iron nears saturation in a positive sense. This would effectively double the total flux change and, consequently, double the energy obtainable from a machine of given size. This can be accomplished by adding "bucking coils" within the orbit, as depicted in Fig. 16b. The bucking coils are wound in opposite sense to the main coils, the direct-current-produced fluxes canceling in the central core but adding at the orbit. A blocking filter must be placed in the bucking coil circuit to prevent the flow of induced current when the main coils are energized with alternating current. The time variation of the magnetic fields is shown in the adjoining plot. This scheme is known as *field biasing*.

Kerst's new betatron is depicted schematically in Fig. 16c. Here the magnetic circuits for the guide field and accelerating flux are separated. Instead of field biasing he employs what is termed *flux biasing* to permit electron acceleration during the swing of the central flux from a high negative to positive value. The negative flux bias is provided by direct current through the distributed bias windings. The accelerating flux is supplied by a unidirectional current pulse through turns *C*. The same pulse through *A* and backwound coil *B* produces the requisite guide field. A unidirectional current pulse is mandatory here as opposed to alternating current, since the central iron is initially biased almost to saturation and the negative half of an alternating-current cycle would seriously saturate the core.

Of particular interest here is the elimination of the air gap in the central core. In a conventional betatron this gap is essential to maintain the 2:1 condition discussed earlier.

In this machine one adjusts the series guide field coils *A* and *B* and flux coil *C* so that the 2:1 condition is obeyed when the voltage across *A* plus *B* is the same as that across *C*. The two sets of coils are then connected in parallel. Thus any tendency to upset the desired condition will induce a correcting current which automatically restores balance. This concept is descriptively titled *flux forcing*. By elimina-



tion of the central gap one achieves a marked reduction in the power and iron required to generate the required accelerating flux.

In the energy range beyond 300 Mev a serious limitation must be faced. This involves the radiation of electromagnetic energy by the electrons. Whenever any body moves in a circular orbit it experiences a radial acceleration  $v^2/r$ . Classical electrodynamics demands that an accelerated charge radiate energy. Thus the revolving electrons are radiating throughout the entire acceleration at a rate which turns out to be proportional to the fourth power of their energy. For heavy ions this effect can always be neglected. Even for electron energies below 100 Mev the effect is not large enough to be serious, but at higher energies it becomes appreciable, thanks to the fourth-power energy dependence. For example, the radiation at 300 Mev is 81 times that at 100 Mev. Once the radiative effect is of consequence, the electrons no longer remain at the equilibrium radius but start spiraling inward. To understand this phenomenon let us agree that the flux change within the orbit has been sufficient to produce 350-Mev electrons. This means the field at the orbit will be such as to keep electrons of this energy coursing the same path. But if the electrons have radiated away 35 Mev of energy they actually possess but 315 Mev. Hence the orbit field will be proportionately too strong, with the result noted above. Kerst finds it possible to compensate for this by imposing a correcting flux pulse at the orbit. Nevertheless it is believed that 500 Mev represents the maximum energy attainable with a conventional betatron.

## The cyclotron

This extraordinarily successful device employing the principle of multiple acceleration is the invention of Lawrence at the University of California. For this work he was awarded the Nobel prize in physics for 1939. Deuterons of 22 Mev and alpha particles of 44 Mev have been produced by this machine. The mammoth new frequency-modulated cyclotron at Berkeley produces deuterons of 190 Mev, protons of 350 Mev, and alpha particles of 380 Mev. The recently completed Nevis (Columbia) frequency-modulated cyclotron turns out protons of 385 Mev.

The heart of a conventional cyclotron consists of a large circular vacuum chamber, inside which are placed two hollow semicircular electrodes called "dees" because each is shaped like a capital D. These dees are situated with their straight edges parallel and about an inch apart, the unit forming an approximate circle when viewed from



above. Figure 17 shows a view of the chamber. The dees are coupled to a potent source of radio-frequency voltage, usually through the intermediary of a coupling link known as a quarter-wave line. This line matches the impedance of the radio-frequency output to that of the dees, thus permitting optimum power transfer. The dees form the high-voltage end of the quarter-wave line. Ions are produced at the center of the chamber between the dees either by fast electrons from an open filament or by an enclosed capillary arc. The whole

chamber is placed between the pole faces of a large electromagnet capable of producing a field in the neighborhood of 16,000 gauss.

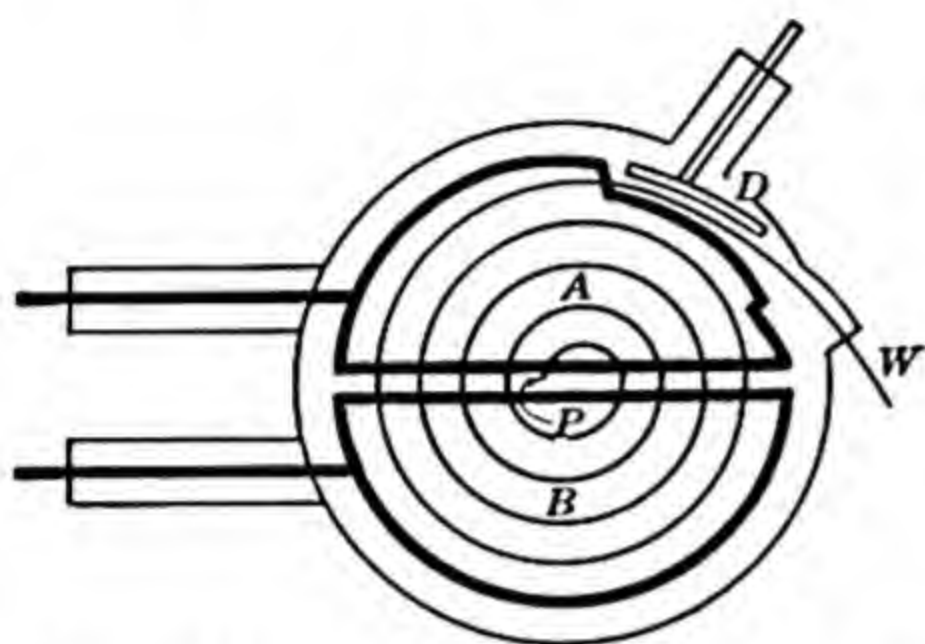


FIG. 17. Cyclotron vacuum chamber. The ion, starting at *P*, follows a spiral path under the influence of successive accelerations across the gap between *A* and *B*, the two dees, and a magnetic field perpendicular to the plane of the motion. It is deflected by the deflector plate *D* and can emerge through a window *W*.

Operation is as follows: The chamber is evacuated to a pressure of approximately  $10^{-6}$  mm of mercury. Gas of the atoms to be accelerated is now introduced into the chamber. As just noted, a cone of ions is formed at the center of the chamber. Let us focus our attention on one of these ions at the moment it is formed and follow its subsequent motion.

Assume that at the moment it is formed at *P* (Fig. 17) dee *A* is at the negative voltage peak of the radio-frequency cycle and hence dee *B* is at its positive peak. The ion, being positively charged, will be drawn over to *A* and will pass into the hollow, field-free interior, acquiring a small velocity. Once inside the dee, the ion is unaware of the charge or change of charge on that dee. However, the magnetic field does act on the ion, exerting a force on it at right angles to its direction of motion. The mathematically minded reader will deduce immediately that this force will cause the ion to move in a circular path. If by the time the ion has completed a semicircle the dees have reversed charge, the ion will again be accelerated across the gap and enter the interior of *B*. Each time it passes from within one dee to the other the ion gains an increment of kinetic energy equal to its charge multiplied by the momentary voltage difference between the two dees. With a high-power radio-frequency supply this difference may be as high as 250,000 volts at the peak of a cycle.



One may calculate the radius of path at any moment by equating the magnetic force on the ion to the mass times acceleration. In cgs units the force can be shown to be given by

$$\frac{Hev}{c} \quad (7)$$

and the mass times acceleration is

$$\frac{mv^2}{r} \quad (8)$$

Here  $H$  is the magnetic field strength in gauss (emu);  $e$  is the effective charge on the ion (esu);  $v$  is the linear velocity of the ion;  $r$  is the radius of curvature of the ion path;  $m$  is the mass of the ion;  $c$  is the velocity of light.

Since by definition the angular velocity  $\omega$  is  $v/r$ , mass times acceleration can also be written

$$m\omega^2 r \quad (9)$$

Combining expressions 7, 8, and 9 we find

$$r = \frac{mvc}{eH} \quad (10)$$

$$\omega = \frac{eH}{mc} \quad (11)$$

The fundamental theory of the cyclotron, frequency-modulated cyclotron, synchrotron, and proton synchrotron is contained in equations 10 and 11. Applying these to the cyclotron we observe from equation 10 the greater the velocity, the greater the radius of path the ion traverses. Thus as the ion gains energy (and consequently velocity, since  $\text{K.E.} = \frac{1}{2}mv^2$ ) through continued accelerations between dees, it travels in ever-widening circles until it reaches the exit slit in dee A where it is pulled to one side by a deflector plate  $D$  charged to a negative potential of some 50,000 volts. Furthermore equation 10 tells us that, since all ions passing through the deflecting system have the same radius, they have the same velocity (assuming  $e$  and  $m$  the same for all) and consequently the same energy.

Equation 11 informs us that the angular velocity of the ions is independent of the linear velocity. In other words, *an ion requires the same time to complete one revolution whether moving near the center of the chamber or close to the periphery.* This rather unexpected



result means that if the magnetic field is adjusted so that the ion returns to the dee gap on its initial half-circle to find the charges on the dees reversed it will continue to do so for each succeeding half-circle. Physically this condition that the time of one revolution be the same, no matter what the diameter of the circular path, can be understood by realizing that the faster an ion moves the farther it must go to complete the circle of greater radius. When the time for one half-circle is the same as that for one half-cycle (radio-frequency), the condition is spoken of as

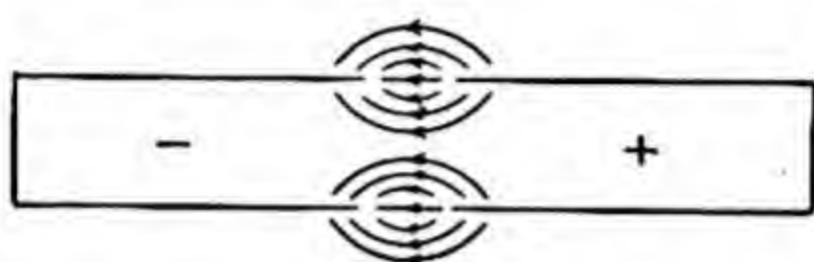


FIG. 18. Electrostatic focusing in the cyclotron. As the ion crosses the gap it gains speed and hence spends less time in the neighborhood of the gap. This means that the electrostatic force tending to drive the ion in to the median plane before acceleration is more effective than that tending to drive it away after acceleration. This produces a net focusing effect which is operative in the early stages of acceleration.

resonance.

In a large cyclotron, even with a peak potential difference of 250,000 volts between the dees, a deuteron must make about 50 revolutions within the chamber before it reaches the final energy of 22 Mev. This requires that the ion travel over 100 meters from the time it is formed until it reaches the target. Although this is a far cry from the distance traveled in a betatron, at first sight it might seem as though only a minute fraction of those starting out could survive such severe solid-angle limitations. However, two happy circumstances prevent this fact from having more than a slightly destructive effect. The first of these, electrostatic focusing (see Fig. 18), is identical to that present in an ordinary accelerating tube which was discussed previously. Since the voltage across the dees is alternating there is one basic difference between this case and that of the conventional accelerating tube where the voltage difference is constant. Just as with the linear accelerator, ions entering the dee gap while the voltage is increasing will experience a net defocusing action. Consequently only those ions crossing the gap while the voltage is decreasing will be focused into the median plane and thus appear in the final beam. As the amount of electrostatic focusing is proportional to the ratio of the velocity across the first half of the gap to that across the second half, and as the energy increment is constant, this type of focusing will lose its effectiveness after a few revolutions of the ion.

During the later revolutions magnetic focusing is mainly responsible for converging the beam. This phenomenon may be visualized by the



aid of Fig. 19. The magnetic lines of force between the pole faces are shown. Toward the periphery the field has a sizable horizontal component. Above the median plane this horizontal component is directed inward; below, it is outward. By the motor rule it is clear that an ion moving clockwise as seen from above, and either above or below the median plane, will be forced back toward the middle by this horizontal field component. Although smaller in actual magnitude, magnetic focusing probably has a greater total effect than its electrostatic counterpart, since it is in operation throughout the whole circular path whereas the electrostatic is operative only in the gap between the dees. So effective is the focusing in a cyclotron that the beam, after passing through the deflecting system, usually covers an area of not more than 1 sq cm.

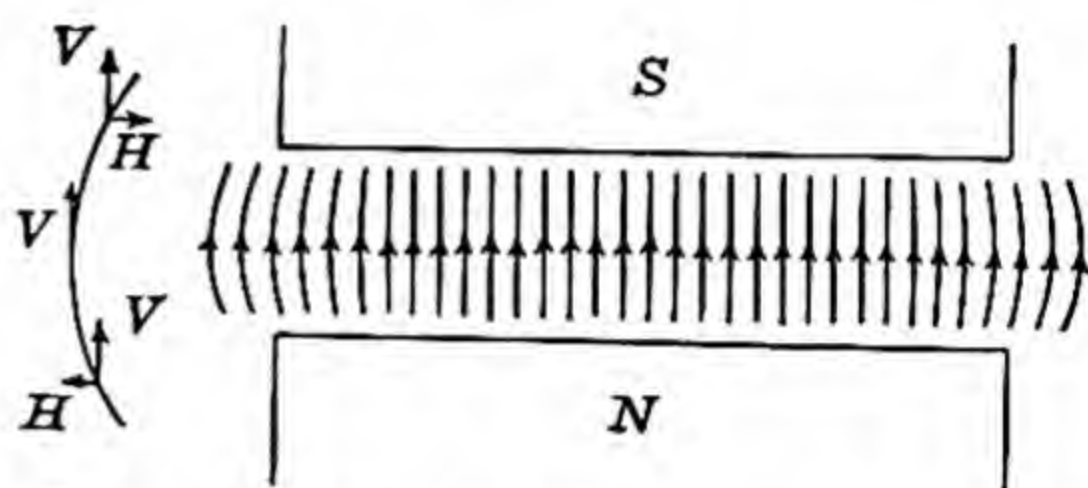


FIG. 19. Magnetic focusing in the cyclotron. The curvature of the field near the edges of the pole pieces means the existence of components of the field in the horizontal plane as shown. These components tend to keep the beam in the center.

This feature of magnetic focusing, although practically indispensable for successful operation, becomes impossible of fulfillment in a conventional cyclotron when very energetic beams are desired. This is on account of the relativity change in mass of the particle as its velocity approaches that of light. Whereas a 1-Mev deuteron has an increase in mass over the rest mass of less than 0.1 per cent, a 200-Mev deuteron has a mass 11 per cent greater than its rest mass. A glance at equation 11 shows that, if we are now to keep the frequency of rotation fixed, the magnetic field must increase as the ion moves outward, else the ion will get out of step with the radio-frequency field. There is no great difficulty in securing a radially increasing magnetic field experimentally, but when it does exist the favorable condition of magnetic focusing is lost, for the horizontal components become reversed and the beam is defocused.

Nevertheless just prior to World War II Lawrence started construction of a 184-inch diameter cyclotron, hoping that through the use of radio-frequency dee voltages in excess of a million so few revolutions of the particles would be required that the defocusing occasioned by a radially increasing magnetic field could be tolerated. Whether or not this brute force scheme would have worked we will never know because while construction was at a standstill during the



war McMillan in the United States and Veksler in Russia independently showed that a controlled variation of certain phase-stable orbits could be utilized to provide a simpler and far neater solution to this problem. In fact this general concept applies equally well to the acceleration of heavy particles and electrons and is so powerful that one is now able to design machines in the Bev region with full confidence of success.

To gain an understanding of this idea let us consider what would happen in the 184-inch cyclotron if we paid the matter of increasing

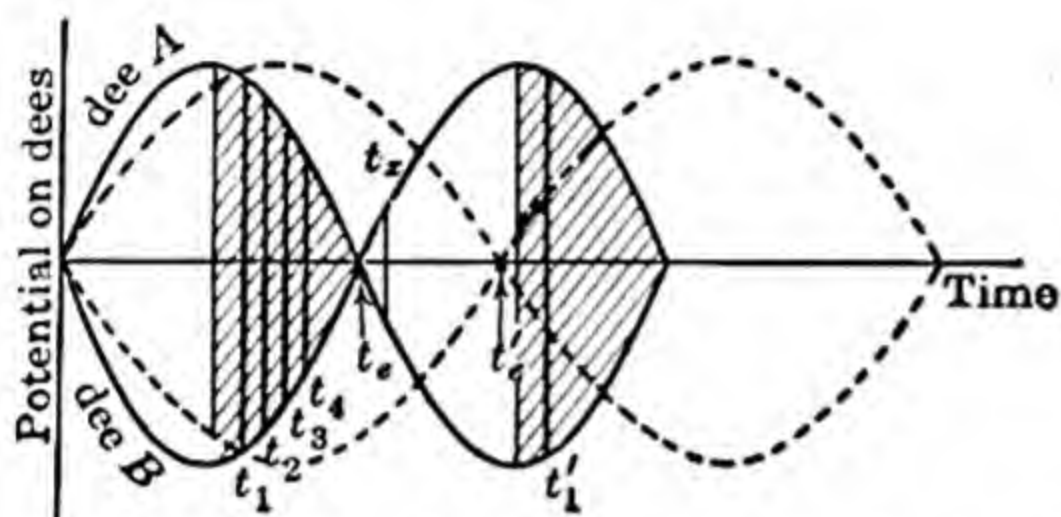


FIG. 20. Graph illustrating effect of increased particle mass on resonance conditions in a cyclotron. One complete cycle of the radio-frequency field is shown. Successive cycles may be imagined superposed on the first. Particle crossing initially at  $t_1$  crosses at  $t_2, t_3, t_4$ , etc., as its mass increases until it is trapped into equilibrium orbit at  $t_e$ . Slow decrease in field frequency as indicated by the dotted lines increases equilibrium energy adiabatically. In this way energies beyond 200 Mev may be obtained. This is the principle of the frequency-modulated cyclotron.

mass no heed. Adopting a constant value of  $H = 15,000$  gauss we find from equation 11 that the oscillator frequency for deuterons would be 11.5 megacycles. Referring to Fig. 20 it is clear from our previous discussion that those particles crossing the dee gap during the shaded portion of the first half-cycle will be focused into the median plane. It will now be shown that, contrary to the linear accelerator case, particles initially passing the accelerating gaps during the shaded part of the radio-frequency cycle also possess phase stability. Consider a particle crossing at time  $t_1$ . So long as its velocity is well below that of light its mass will not change and it will continue to reach the gap at a corresponding time during succeeding cycles.

But once its mass begins to grow due to increased *linear* velocity, equation 11 reminds us that its *angular* velocity will decrease. Thus it will require slightly longer to complete a circle and will reach the gap at a later time  $t_2$ . It will still be accelerated under these conditions, but the voltage kicks become smaller. The relative times of arrival during subsequent cycles will be  $t_3, t_4$ , etc. Finally the time will come when the ion crosses the gap at  $t_e$ . But at this instant there is no voltage difference between the dees and the ion receives no kick. If for some reason it happened to cross at a still later time  $t_x$  it would be decelerated since the field is here reversed. This would effectively



decrease  $v$  and  $m$  and increase  $\omega$ , and the next gap crossing would be shifted back toward  $t_e$ . In other words, the  $t_e$  condition represents an equilibrium orbit in which the particle is imprisoned. Consequently under the conditions stated above no beam would ever reach the deflector plate but at some intermediate path radius there would be a circulating beam of deuterons trapped in this equilibrium orbit. What Veksler and McMillan effectively showed was that if the energy associated with this orbit were changed adiabatically by varying either the magnetic field or the oscillator frequency the particles circulating in this orbit would automatically keep in step with the change (provided the change were slow compared to the time required for the particle to make a revolution) and thus particles could be accelerated to very high energies irrespective of any change in their mass.

Three new types of accelerator embody this idea: (1) the frequency-modulated cyclotron, (2) the synchrotron, and (3) the proton synchrotron. Since each accomplishes this end in a different manner we shall discuss their operation in some detail.

### Frequency-modulated cyclotron

Suppose in the above situation we decrease the oscillator frequency by a small amount. This is indicated by the dotted lines of Fig. 20. The  $t_e$  particles now find an accelerating field waiting. They gain energy and move in a larger circle, their angular velocity decreases slightly, and soon they have an arrival time hovering around  $t_e'$ . This trick can be repeated over and over. In practice the frequency is decreased smoothly, the equilibrium orbit expanding to larger and larger radii with the ions following obediently, attaining higher and higher energies. Finally the ion pulse reaches the periphery of the dee where it may be directed against a target. We must now return the frequency to its original value, trap another bunch of ions, and repeat the trick of frequency variation. By repeating this process over and over we are able to produce a stream of ions with energies undreamed of in the days of regular cyclotrons. At the University of California the radiofrequency has been modulated up to 2000 times a second by means of a rotating condenser in the dee line. This means we get only 2000 ion bursts per second instead of the 20 million obtained from a conventional cyclotron. The picture is not quite so dark because the ion bursts in the frequency-modulated machine are larger. Still it is calculated that a frequency-modulated cyclotron will produce time average beams only 5 per cent of those from a normal cyclotron.



However, one willingly sacrifices numbers in order to secure the much greater energies.

It will be instructive to calculate the final mass and energy of the deuterons in the example discussed above. To do this we must write equations 10 and 11 in their relativistic form, i.e., replace  $m$  by  $m_0/\sqrt{1-\beta^2}$  where  $m_0$  is the rest mass of the particle and  $\beta = v/c$ . Equation 10 becomes

$$r = \frac{m_0 c^2 \beta}{\sqrt{1-\beta^2} e H} \quad (12)$$

Solving for  $\beta^2$

$$\beta^2 = \frac{(Her)^2}{(m_0 c^2)^2 + (Her)^2}; \quad \beta = \frac{Her}{\sqrt{(m_0 c^2)^2 + (Her)^2}} \quad (13)$$

Equation 11 becomes

$$\omega = \frac{\sqrt{1-\beta^2} e H}{m_0 c} \quad (14)$$

The final ion radius in the 184-inch California cyclotron is 81 inches = 206 cm. Putting this value in equation 13 and assuming  $H = 14,300$  gauss at the dee edge (the field is made to decrease radially to improve magnetic focusing, which in turn eliminates the need for electrostatic focusing and permits the use of a single dee, a welcome simplification), we find  $\beta^2 = 0.186$  or  $\beta = 0.432$ . Thus the deuterons are traveling at almost half the velocity of light. Their mass is now

$$m = \frac{m_0}{\sqrt{1-\beta^2}} = \frac{m_0}{\sqrt{1-0.186}} = 1.107 m_0$$

or 10.7 per cent greater than the rest mass. According to the Einstein law the *total* energy possessed by a moving particle is  $mc^2$ . This can be broken down into rest mass energy ( $m_0 c^2$ ) and kinetic energy. Thus

$$mc^2 = m_0 c^2 + \text{K.E.} \quad (15)$$

and

$$\text{K.E.} = \frac{m_0}{\sqrt{1-\beta^2}} c^2 - m_0 c^2 = m_0 c^2 \left( \frac{1}{\sqrt{1-\beta^2}} - 1 \right) \quad (15a)$$

Putting the appropriate values in equation 15a leads to a figure of 200 Mev for the kinetic energy of the deuterons.

To find the amount of frequency modulation needed we use equation 14. At the start of acceleration  $\beta = 0$  and equation 14 becomes



$$\omega_0 = 2\pi f_0 = \frac{eH}{m_0c}$$

(16)

Dividing equation 14 by equation 16 gives

$$\frac{f}{f_0} = \sqrt{1 - \beta^2} = 0.905$$

Thus the frequency must decrease around 10 per cent or from 11.5 to 10.4 megacycles. A decrease of 16 per cent is actually used at the University of California in order to keep on a fairly straight portion of the frequency-time curve. Helium ions and protons have also been accelerated in the 184-inch machine. The following table lists the pertinent information on these particles calculated as above. A comparison between the frequency modulated and conventional cyclotrons is given in Fig. 21.

PARTICLE	H (gauss)		r (cm)	f <sub>0</sub> (mc)	β	m	K.E. (Mev)	f/f <sub>0</sub>	f (mc)
	Center	Edge							
Deuteron	15,000	14,300	206	11.5	0.43	1.11m <sub>0</sub>	200	0.905	10.4
Helium ion	15,000	14,300	206	11.5	0.43	1.11m <sub>0</sub>	400	0.905	10.4
Proton	15,000	14,300	206	23.0	0.69	1.38m <sub>0</sub>	355	0.726	16.7

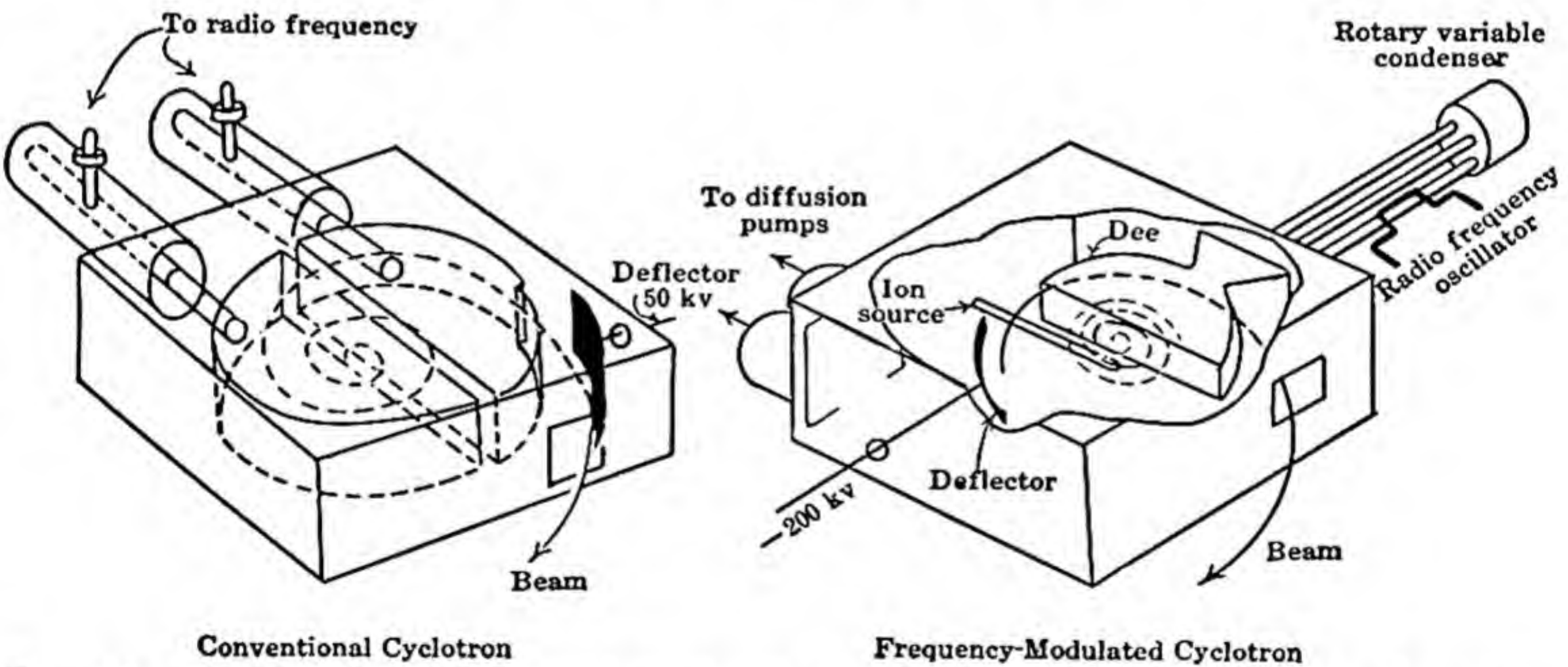


FIG. 21. Operation of conventional cyclotron contrasted with that of a frequency-modulated cyclotron.

	Conventional Cyclotron	Frequency-Modulated Cyclotron
Diameter	60 inches	184 inches
Number of dees	2	1 (grounded tank replaces other dee)
Maximum energy (deuterons)	22 Mev	200 Mev
Maximum energy (helium ions)	44 Mev	400 Mev
Magnetic field	15,000 gauss	15,000 gauss
Radio frequency	Constant	Variable
Radio-frequency voltage on dee	50,000-200,000 volts	15,000 volts
Radio-frequency power	100 kilowatts	18 kilowatts
Total revolutions for ion	50-200	10,000
Time for ion acceleration	10 microseconds	1000 microseconds
Beam current	100 microamperes	0.6 microampere



### The synchrotron

As has been stated, the principle of phase stability also governs the operation of the synchrotron. However, since the synchrotron is designed to produce high-energy electrons as opposed to protons, deuterons, and the like, there are differences between the synchrotron and the frequency-modulated cyclotron in construction and operation. A 300-Mev electron possesses a mass of  $600m_0$  (electron rest mass) whereas a 300-Mev proton has a mass of but  $1.38m_0$  (proton rest

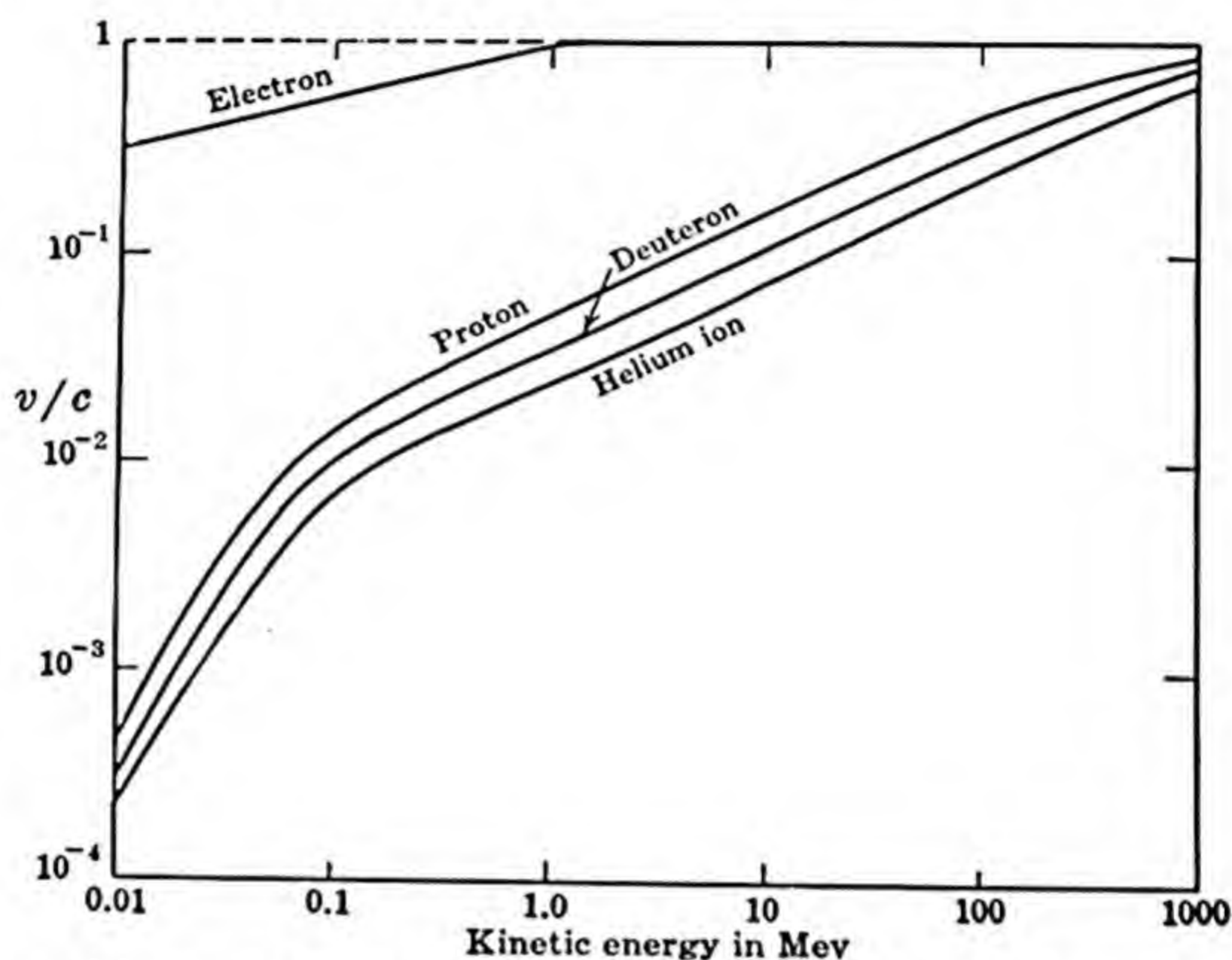


FIG. 22. Logarithmic plot showing the variation of  $v/c$ , i.e.,  $\beta$ , for the four charged projectiles commonly used in nuclear bombardment work. Of particular interest is the fact that for energies in excess of 1 Mev the electron moves essentially at the velocity of light.

mass). Inasmuch as the relative frequency variation in an equilibrium orbit accelerator must be equal to the relative mass change this would call for a 600-fold change in frequency for the 300-Mev electron, a feat not currently possible. However, equation 11 informs us that increasing  $H$  would be just as satisfactory for maintaining resonance as decreasing  $f$  ( $\omega = 2\pi f$ ), and this is just what is done in the synchrotron.

Another difference in the two devices lies in the relative magnet geometry. Figure 22 plots the variation in velocity with energy for electrons and three heavier particles. If it is recalled that the path radius is  $\propto (mv/H)$ , it is clear that the orbit of a proton, deuteron, or helium ion in a frequency-modulated cyclotron shows a large change



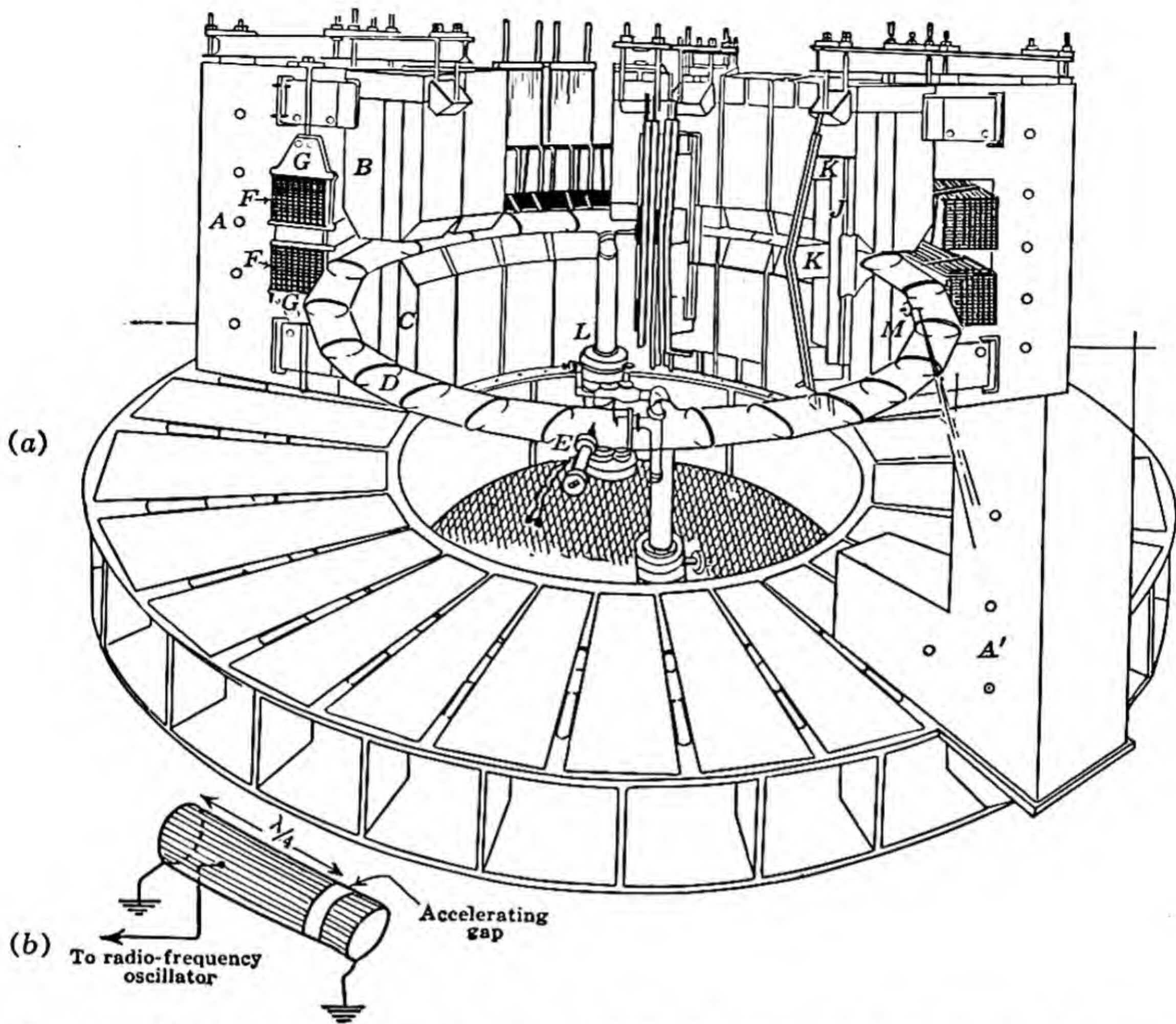


FIG. 23. (a) Cutaway sketch to show the main construction features of a synchrotron. A and A' are views of laminated iron C-shaped magnet sections which are joined together to supply the necessary field. The magnet is energized by a 60-cycle current through coils F which are a part of a resonant LC circuit. The flux bars J bridge the open face of the magnet and carry the magnetic flux which produces the initial betatron acceleration. The doughnut accelerating tube is made from many elliptical Pyrex tube sections D joined together with rubber gaskets. The tube is evacuated by diffusion pump L. E is the electron gun and M the target. (*The Cornell Engineer*, December 1948, p. 11.) (b) One section of Pyrex tube may be silver coated as shown to form the radio-frequency resonator (not pictured). Longitudinal scratches in the coating prevent eddy currents which would distort the field within the tube. Voltage multiplication at the acceleration gap may be obtained by feeding in the radio frequency near the grounded end of the quarter-wave section.



in radius over the full acceleration. Consequently a large frequency-modulated cyclotron magnetic field area is mandatory. On the other hand, if  $H$  varies with  $m$  so that  $m/H$  is constant, an electron will experience practically no change in orbit radius in going from, say, 2 Mev to 300 Mev. Furthermore it should be remembered that a betatron accelerates electrons from low energy to the Mev range at a constant radius. Hence if we could devise things so that the synchrotron operated initially as a betatron it should be possible to get by with a lightweight ring-shaped magnet. This is most desirable because the magnet normally represents a major cost item in the construction of a circular accelerator. This idea has proved quite successful, and most synchrotrons built or being built operate in this manner. In fact, a synchrotron differs from a betatron in only two main features: (1) the synchrotron requires only a slight amount of magnetic flux inside the electron orbit, and (2) it has a radio-frequency electrode in the doughnut which serves to boost the electrons to high energy after the betatron action has spent itself. Figure 23 illustrates the general appearance of a synchrotron. Operation is as follows: A pulse of 30- to 60-Kev electrons is injected into the doughnut as the magnetic field passes through zero. These electrons are quickly accelerated to 2 Mev by the betatron principle, at which point the flux bars become saturated even though the field through the doughnut continues to rise. The radio-frequency oscillator is now turned on, and the electrons receive an increase in energy each time they cross the radio-frequency electrode gap, their velocity remaining constant at  $c$  but their mass becoming greater. Since the time per revolution should not vary, the oscillator frequency is likewise constant. To visualize how the principle of phase stability functions here we refer again to equation 11 which can be written

$$\omega = \frac{eH}{mc} = \frac{eHc}{mc^2} = \frac{eHc}{E} \quad (17)$$

Here  $E$  is the kinetic energy of the electron, provided we ignore its  $\frac{1}{2}$ -Mev rest mass energy. If  $H$  increases faster than  $E$ ,  $\omega$  will increase (the electron moving to a smaller orbit), and the electron will reach the gap early next time around and receive a bigger kick (Fig. 20). If the rise in  $H$  is too slow just the reverse condition obtains. In any event phase stability will assist in maintaining  $H/E$  constant.

If the doughnut has a radius of 100 cm the time for one revolution will be  $2\pi(100)/c$  which must also be the time for one radio-frequency oscillation. Thus



$$f = \frac{1}{T} = \frac{c}{2\pi(100)} = 47.7 \text{ megacycles}$$

Putting this value in equation 17 and assuming a final magnetic field of 10,000 gauss we obtain for the final energy

$$E = \frac{(4.81)(10^{-10})(10,000)(3)(10^{10})}{(2\pi)(47.7)(10^6)(1.6)(10^{-6})} = 300 \text{ Mev}$$

Several 300-Mev synchrotrons are now operating successfully. Since the principle of phase stability is able to compensate for energy radiated by the electrons so long as the amount radiated is small compared to that gained per turn, it is quite possible to envisage such machines operating in the Bev area if the demand for such energetic electrons or gamma rays arises. One such project is currently under way at the California Institute of Technology under the auspices of the Atomic Energy Commission. Otherwise, however, this energy region is being assaulted by a heavy particle accelerator which is a combination frequency-modulation cyclotron-synchrotron machine and which answers to the name proton synchrotron, bevatron, or cosmotron, depending on the scientific circle in which one moves.

### Proton synchrotron

Primary cosmic rays are predominantly protons with energies in the Bev range, and the phenomena they cause are intriguing. At present they represent our most productive source of knowledge concerning nuclear forces. Thus a machine which would produce 5- to 10-Bev protons in quantity has been the dream of nuclear scientists for years.

At first sight the engineering problems associated with a 6-Bev proton accelerator stagger the imagination. If we write equation 10 as  $r = mc^2\beta/eH$  and substitute equation 15 for  $mc^2$  and equation 13 for  $\beta$ , we get

$$r = \frac{\sqrt{(K.E. + m_0c^2)^2 - (m_0c^2)^2}}{eH} \quad (18)$$

Substituting in equation 18 values appropriate to a 6-Bev proton (assuming  $H = 15,000$  gauss) we find that  $r = 1530$  cm or 50 feet. Not even the most enthusiastic cyclotroneer would seriously consider a frequency-modulation cyclotron magnet 100 feet in diameter. So we must try some other approach. The synchrotron scheme involving a thin ring magnet would be most attractive here since even a diame-



ter of 100 feet would not be insurmountable. But there is one big difficulty to be faced. The synchrotron electrons move with a constant velocity  $c$  throughout the whole radio-frequency acceleration, whereas even at 1 Bev a proton is appreciably below  $c$  and it is still accelerating. However, this problem was solved in the frequency-modulated cyclotron by varying the oscillator frequency. So it all boils down

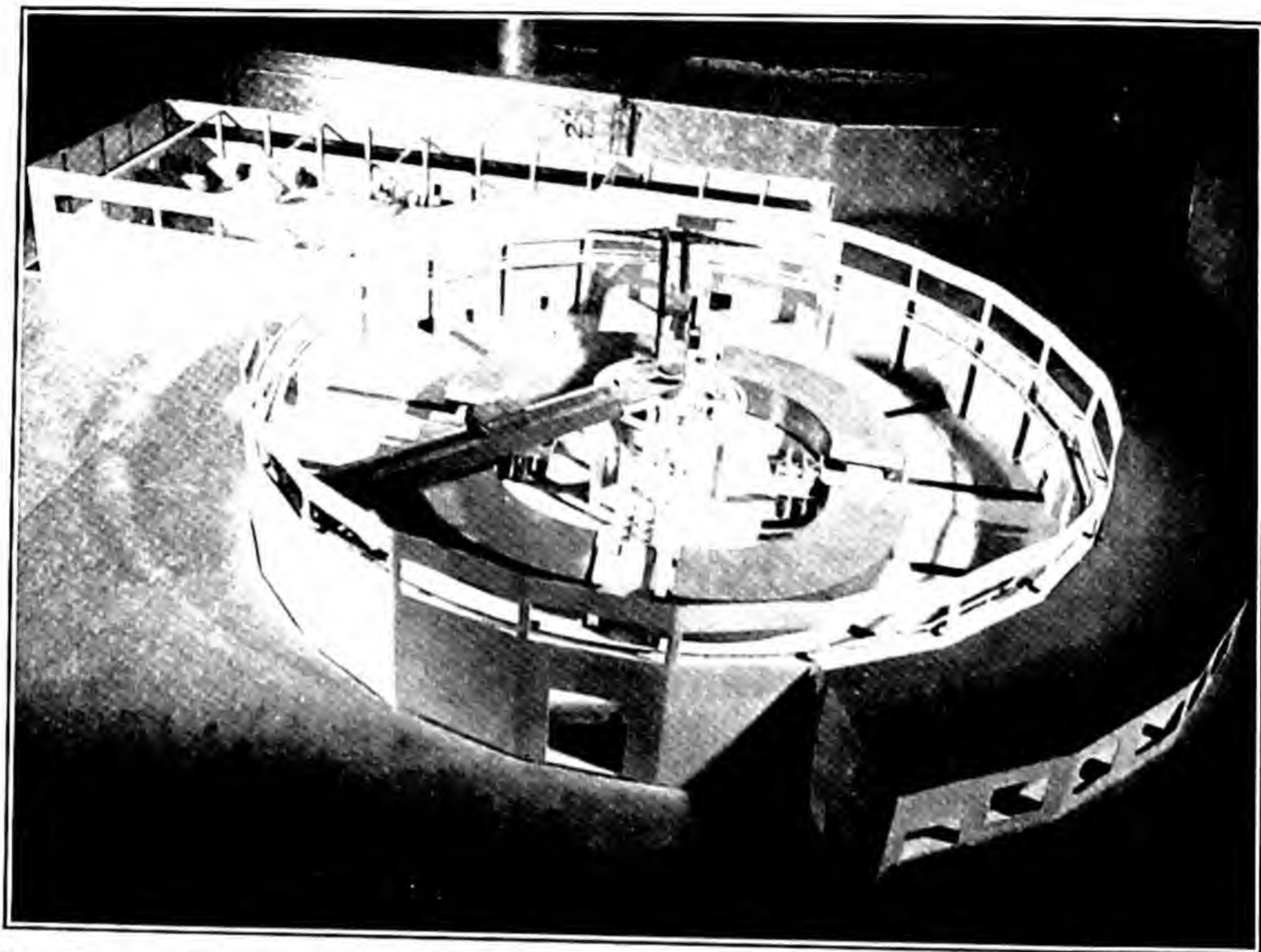


FIG. 24a. The giant bevatron now under construction at the University of California, financed by the Atomic Energy Commission. It is anticipated that this instrument will produce protons with energies as high as 6 Bev. A somewhat smaller proton synchrotron of this type is being built at the Brookhaven National Laboratory. (Courtesy Radiation Laboratory, University of California, and U. S. Atomic Energy Commission.)

to the fact that a 6-Bev proton accelerator is technically possible if we are willing and able to vary both magnetic field and frequency.

Three machines designed to operate in this manner are a 1.3-Bev instrument at Birmingham University in Great Britain, one rated for 3-Bev at the Brookhaven National Laboratory, and a 6-Bev machine at the University of California. The latter two are being financed by the Atomic Energy Commission. Figure 24a pictures a scale model of the bevatron and gives an idea as to how it will look when completed. Some vital statistics on the California bevatron



may be of interest. The magnet will consist of four 90-degree annular segments disposed so that the proton orbits are quarter-circles connected by straight sections. The orbit radius will be roughly 48.5

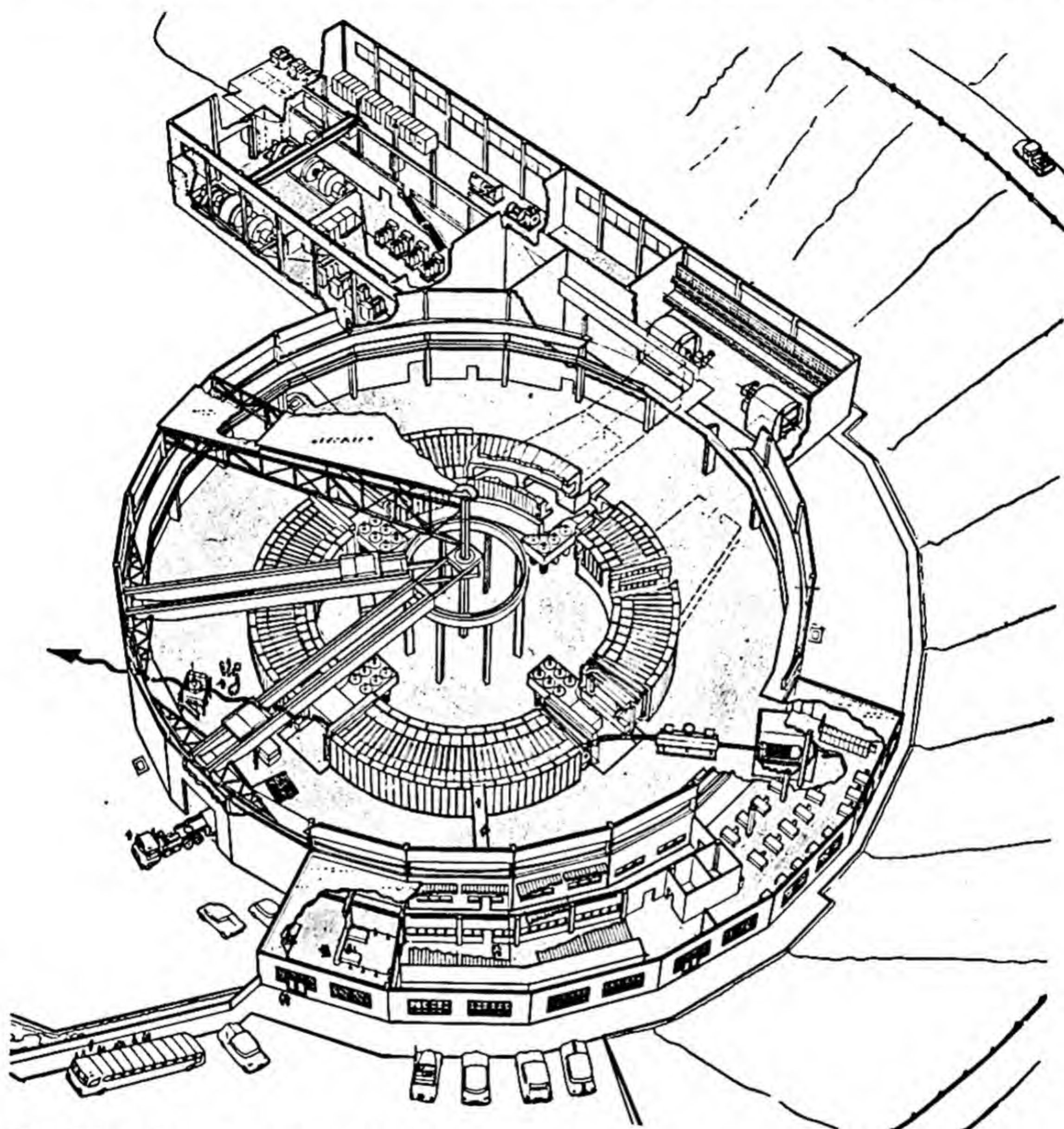


FIG. 24b. Cutaway sketch of the bevatron, showing the disposition of the various components. The protons will be given an initial acceleration in the linear accelerator and then turned into the racetrack-shaped accelerating tube where they will be speeded up to their final energy. (Courtesy Radiation Laboratory, University of California, and the U. S. Atomic Energy Commission.)

feet, and the length of the straight sections will be 20 feet. The rectangular tube in which the ions move will have usable cross-section dimensions of 24 inches (vertical) by 72 inches (horizontal). Magnet power will be furnished by a motor generator with a large flywheel



attached. When the magnetic field is increasing, energy will be taken from the flywheel and stored in the magnet. While the magnetic field is decreasing, the generator will act as a motor and return energy to the flywheel. In this way one has only to supply enough outside power to make up for the magnetic, mechanical, and  $I^2R$  losses in the system. To initiate operation a pulse of protons, accelerated to 10 Mev in a linear accelerator, will be fired into the bevatron tube when the magnetic field rises to some 300 gauss. The electric field between a pair of curved inflector electrodes will serve to turn the protons so their path is tangent to the bevatron orbit. Radio-frequency power will now be applied to the accelerating electrode, the frequency being 0.4 mc. Both magnetic field and resonator frequency now rise in a controlled manner, phase stability sufficing to keep the protons in synchronism. When the frequency reaches 2.5 mc the magnetic field will have a value of 9800 gauss and the protons will be moving with an energy of roughly 3.7 Bev. Initially it is planned to operate the bevatron at this energy. Later, modifications will be made to allow a maximum magnetic field of 15,000 gauss, which with a higher resonator frequency will permit the attainment of 6 Bev energy. It is estimated that each final pulse will contain about  $10^{10}$  protons, about 5 per cent of those starting the long 270,000-mile circular journey. The acceleration time will be roughly 1.75 seconds, and the machine will be pulsed 10 times per minute. The average beam current will be minute compared to the output of low-energy machines but tremendous in contrast to the average flux of cosmic rays reaching any spot on earth.

The reader may wonder at the confident manner with which we write of something which at the moment (1950) has not come to pass. Our faith is indeed well founded. A quarter-scale model of the bevatron has already been constructed at California, and it operated precisely as predicted. In betting parlance the bevatron certainly looks like a "sure thing."

The machines described in this chapter represent the main artillery with which scientists are currently mounting their attacks on the nucleus. Space does not permit a discussion of several other devices such as the racetrack (a modified synchrotron), the nonferromagnetic synchrotron (wherein the requisite magnetic fields are produced by air core coils carrying tremendous momentary currents), the microtron (an electron cyclotron), and a number of other instruments that hold promise but have not yet demonstrated their ability to compete



with the foregoing accelerators. Perhaps in the future they will take their place on the firing line. With such formidable weapons at our disposal it seems a certainty that the nucleus must soon yield up its innermost secrets.

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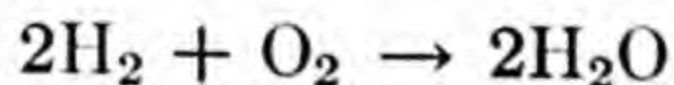


## 5. Transmutation

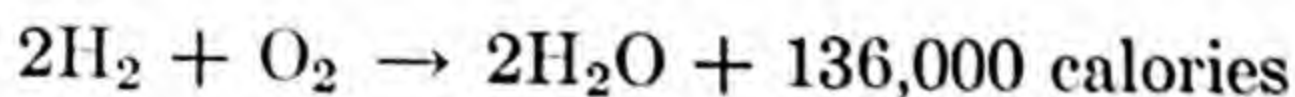
Today over a thousand nuclear reactions are known, an astonishing development since the first artificially induced nuclear reaction in 1919. The long-awaited flower of alchemy has broken into full blossom in two decades. This abundance of reactions calls for some systematization, correlation, and explanation; in fact, it calls for the development of the new subject of nuclear chemistry, and in this chapter we intend to present the rudiments of that subject.

In the first chapter we introduced the idea of transmutation and illustrated it by describing some of the pioneer work of Rutherford, Blackett, Chadwick, Curie, and Joliot, who employed natural radioactive sources to provide bombarding projectiles. The technique of artificial acceleration described in Chapter 4 and of the chain-reacting pile described in Chapter 11, has put transmutation on a totally different plane, for now in place of alpha particles as primary bombarding agencies we have neutrons, protons, deuterons, and mesons available in overwhelmingly greater numbers. It is not surprising that so many new transmutations have been discovered in so short a time. We intend to discuss transmutations by all these particles, as well as by neutrons and gamma rays, but first we propose to consider a feature of nuclear reactions common to all, and of the greatest importance, namely, *energy relationships in reactions*.

Suppose that we write a reaction in ordinary chemistry:



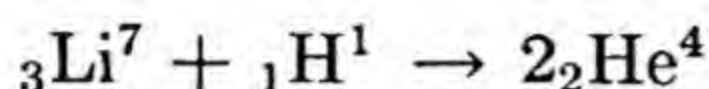
The chemist is not content until he can add to this equation a term expressing the gain or loss of heat in the process, the "heat of reaction." Such an equation then reads



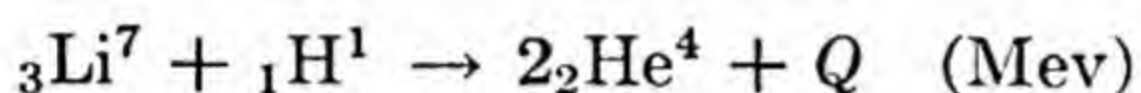
which means that when two gram molecules of hydrogen and one of oxygen combine with water as the final product 136,000 calories are



evolved. Heat may also be absorbed, and the evolution or absorption of heat is described by the self-explanatory terms exothermic or endothermic reaction. If a chemical reaction is exothermic the temperature of the molecules rises; if endothermic, the heat needed is supplied from the molecules by their fall in temperature. Temperature in turn means the energy of motion of the molecules. Now in the same way we may write an equation:



which refers to the bombardment of lithium by protons with the evolution of helium. Like the chemist we are still not satisfied until we add a term analogous to the heat of reaction, a term called (for want of a better) "nuclear energy change," and universally represented by the symbol  $Q$ . This representation has become so widely accepted that "nuclear energy change" and " $Q$  value" are synonymous. The equation then reads



and again we keep the terms exothermic or endothermic for reactions in which  $Q$  is positive or negative, respectively.

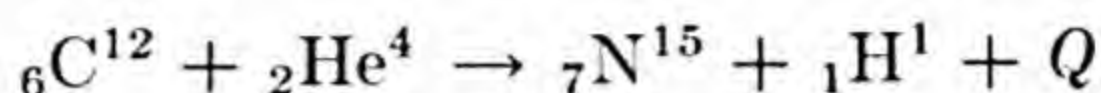
Let us now suppose that  $Q$  is positive, or that in the reaction energy is released. Why is there this release of energy, and where is the seat of the energy whence it came? The answer is similar to the answer in chemistry. The new product is more stable than the two former constituents separately; the particles are more tightly bound together, and this means that in lithium and hydrogen separately we have a surplus of energy over the combination to form two helium nuclei. The nuclei of lithium and hydrogen are supplied by nature with energy to spare—*why* we have yet to discover, but it is so—and our setting off the reaction has liberated the energy that was already available. The energy liberated in a large  $Q$  value is the "atomic energy" that has been discussed so much in popular literature since 1920.

### Balance sheet of mass and energy

The reason why "atomic-energy release" is so sensational a subject is the enormous magnitude involved as soon as we consider these nuclear reactions in terms of their chemical analogues. Thus, when 4 grams of hydrogen and 32 of oxygen combine, 136,000 calories, a fair amount, is released. Yet the combination of 7 grams of lithium and 1 of hydrogen would release roughly 5 billion calories, a stupendously



greater amount.\* This greater release of energy per atom gives the physicist a head start on the chemist, for it means that he can, by a physical measurement to be described, *predict* the energy change in any reaction whatever. He can, for example, predict that the reaction



will have a large negative  $Q$  value and hence will not proceed unless the bombarding alpha particles ( $\text{He}^4$ ) are extremely energetic. This physical measurement is simply *the mass of each of the atoms concerned*. According to the theory of relativity there is a relation between the mass a body possesses and its energy; to the ordinary energy must be added the quantity  $mc^2$ , where  $m$  is the mass and  $c$  is the velocity of light. Ordinarily this term appears equally before and after some change and so may be subtracted out or not even included, but where there is a heavy release of energy in a nuclear reaction *the release is at the expense of mass* and linked to it by the equivalence relation

$$E = mc^2$$

In the years from 1922 to the present day, physicists, notably Aston, Bainbridge, and Dempster, have developed methods of measuring atomic masses with great refinement. Thus it is well known that the atomic weights of most elements are not whole numbers, a fact partially accounted for by the simultaneous presence of isotopes of different atomic weight in the same element. The entire deviation from whole numbers is not due to this cause, for accurate measurements show, for example, that the "atomic weights" of the isotopes  $\text{Li}^7$ ,  $\text{He}^4$ , and  $\text{H}^1$  are 7.01818, 4.00389, and 1.00813, respectively. These atomic weights refer to the masses of a specific number of atoms, the number in a gram atom, and to obtain the actual mass of a single atom of these isotopes we need to divide by the number of atoms in a gram atom. When this is done we find that the mass of a single atom of an isotope of "atomic weight" unity is  $1.66 \times 10^{-24}$  gram. The mass of a single atom of  $\text{Li}^7$  is then  $7.01818 \times 1.66 \times 10^{-24}$  gram. Now, if we consider the balance sheet of mass in the reaction below, where the actual masses of the atoms taking part are written in, we see that mass is not conserved.

\* There is a difference between the nuclear reactions produced by bombardment and chemical reactions as ordinarily thought of. Chemistry is a statistical process of reactions which can go at ordinary temperatures. Except in a few instances, notably in stars and the atomic bomb, this type of statistical process is not involved in nuclear reactions.



$$\begin{array}{ccc}
 \text{Li}^7 & + & \text{H}^1 & \rightarrow & 2\text{He}^4 \\
 \underbrace{7.01818 \times 1.66 \times 10^{-24} + 1.00813 \times 1.66 \times 10^{-24}}_{8.02631 \times 1.66 \times 10^{-24}} & & & & \underbrace{2 \times 4.00389 \times 1.66 \times 10^{-24}}_{8.00778 \times 1.66 \times 10^{-24}}
 \end{array}$$

The two sides do not balance; there is an excess of  $0.01853 \times 1.66 \times 10^{-24}$  on the left-hand side. This is a loss of mass in each individual reaction of  $3.16 \times 10^{-26}$  gram. According to our energy relation  $E = mc^2$ , we therefore expect a release of energy of  $3.16 \times 10^{-26} \times (3 \times 10^{10})^2$  or  $2.76 \times 10^{-5}$  erg *per individual reaction*. Expressed in terms of the energy to accelerate an electron to the same energy this is 17 Mev. We therefore are led to expect as a result of the reaction that each helium nucleus will acquire  $1\frac{1}{2}$  or 8.5 Mev as a result of the reaction, and since to a definite energy of a helium nucleus there is a definite range we expect to find helium nuclei (or "alpha particles") projected with a range of 8 cm in air, the value appropriate to 8.5 Mev. This is accurately verified, and the verification is one of the greatest triumphs of the theory of relativity.

We now have a beautifully compact means of expressing the potentialities of a nucleus for energy evolution—its mass. This fact has given great impetus to the measurement of nuclear masses, and the gradual compilation of a comprehensive table of all nuclear masses is one of the tasks that lies ahead of the physicist. To illustrate the use of masses we give here a shortened table of masses of neutral atoms.\*

H <sup>1</sup>	1.00812	Li <sup>6</sup>	6.01690
n <sup>1</sup>	1.00899	Li <sup>7</sup>	7.01804
H <sup>2</sup>	2.01472	Be <sup>8</sup>	8.00777
H <sup>3</sup>	3.01704	Be <sup>9</sup>	9.01497
He <sup>3</sup>	3.01701	B <sup>10</sup>	10.01605
He <sup>4</sup>	4.00389	B <sup>11</sup>	11.01286

With these the reader can see, for example, that two deuterons may collide with the release of either H<sup>3</sup> and H<sup>1</sup> or He<sup>3</sup> and n<sup>1</sup>. Both reactions are known and are very prolific. He can also see that Be<sup>8</sup> is almost exactly equal to two He<sup>4</sup>'s or two alpha particles. In fact, Be<sup>8</sup> is probably unstable for the reason that it can split up into two alpha particles. An interesting diversion is to use the table to calculate the energy release in any reaction one cares to invent and test whether the transmutation could be made to go.

For convenience in using tables of masses two conversion factors are given here.

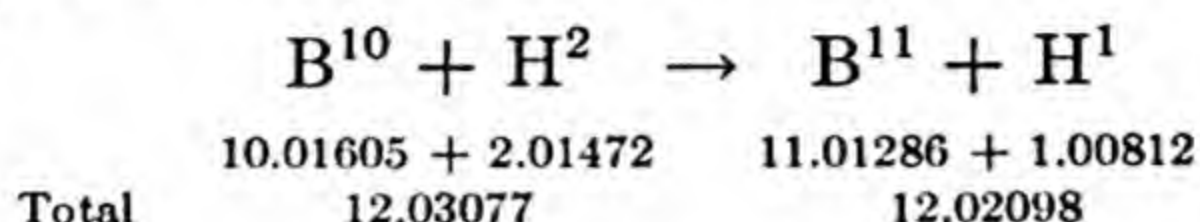
\* A table complete, so far as is now known, is given in Appendix 6.



$$0.0011 \text{ "mass unit"} = 1 \text{ Mev}$$

$$1.6 \times 10^{-6} \text{ erg} = 1 \text{ Mev}$$

Then, for example, the reaction produced when  $B^{10}$  is bombarded by deuterons to give  $B^{11}$  and a proton can be treated thus:



Balance = 0.00979 on the left-hand side, indicating an energy release of 9.1 Mev. This has been observed to be the fact.

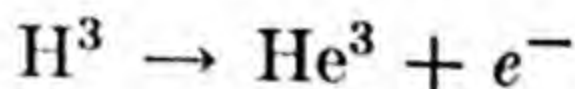
It should be noticed that the masses of *neutral atoms* are given. This is convenient for workers with mass spectrographs and is no trouble to the nuclear physicist. The electrons which must be added to a nucleus to form a neutral atom will always balance out if we

neglect all electrons, with one exception, the emission of a positron. Thus, for example, in the reaction above,  $B^{10}$  has five electrons,  $H^2$  one, making a total of six, while  $B^{11}$  also has five and  $H^1$  one, also totaling six. Or, again, suppose that we consider the radioactive decay of  $C^{14}$ :



$C^{14}$  has six electrons,  $N^{14}$  seven. Now if we take the difference in mass between these neutral atoms we have already included in the

balance sheet the additional electron; the fact that a fast beta particle leaves the  $C^{14}$  and an additional electron is attracted from outside to neutralize the newly formed  $N^{14}$  is immaterial. For greater clarity let us take the very simple example



What actually occurs is indicated in Fig. 1 at A. The dotted lines indicate a vacant space to be filled by an electron. If we consider the change to be as in the figure at B we have included all the particles taking part quite adequately; any difference of mass will then tell us what energy is available. This energy is divided between the fast-moving beta ray and the incoming neutralizing electron, but the latter

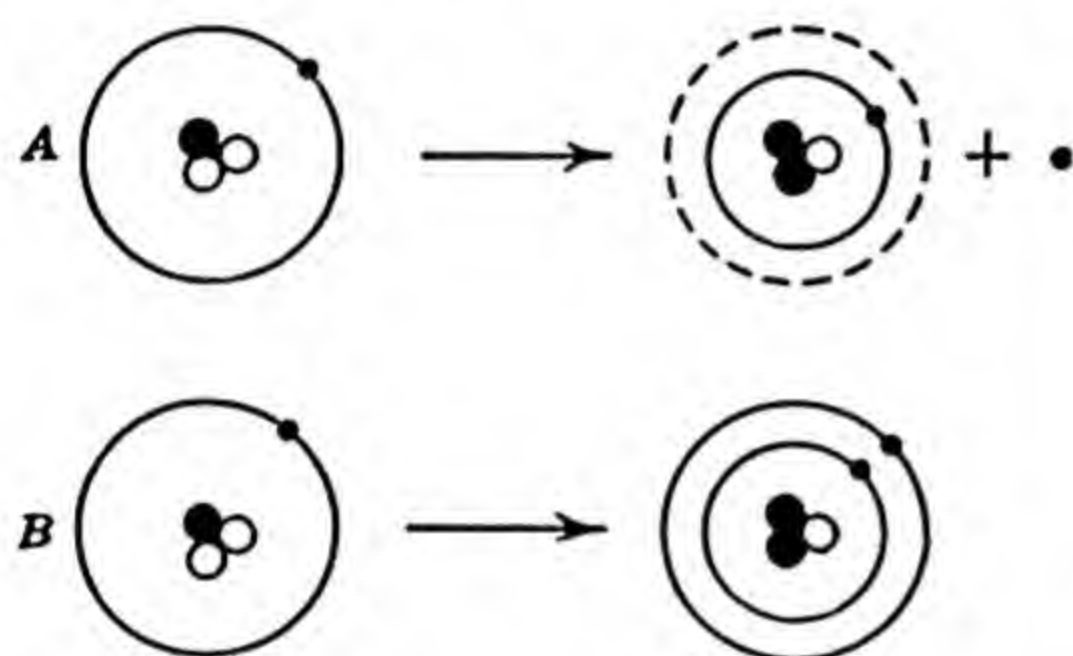


FIG. 1. The use of masses of *neutral atoms* to determine energy changes. The neutral atom formed as indicated in B is equivalent to the actual process in which an electron is evolved and possibly lost as at A.



is a few electron volts only and hence may be neglected. The emission of a positron, however, is different. For example, in the reaction



the nitrogen has to lose first a positron and also an electron in an orbit, or pictorially the process is as shown in Fig. 2. We must therefore add two electron masses or 0.0011 mass unit to the right-hand side or else we shall predict too great an energy for the positron. It is useful to remember that this correction is 1.02 Mev or very nearly 1 Mev.

To summarize about energy relations and atomic masses, we may say that if we consider any reaction we can predict the energy release in million electron volts if we find the difference between the neutral atom masses and use the conversion factor from mass units to million electron volts. If a positron is emitted it must be treated as costing 0.0011 mass unit or 1 Mev extra.

### Nuclear reactions in general

Having seen how a nuclear physicist predicts the energy lost or gained in a nuclear reaction, let us now see how the reactions are actually made to go. We have already mentioned that an establishment equipped with a moderate-sized accelerator has available the following projectiles: protons, neutrons, deuterons, and alpha particles. A chain-reacting pile (or nuclear reactor) makes available very large numbers of neutrons. In addition gamma rays also can cause transmutations. One can therefore readily see that many kinds of combinations are possible and that a complete description of every reaction would need several volumes. To the person who, like the writers, recoils from such a massively detailed subject we can offer some solace. In the first place *almost any energetically possible reaction can be made to go*. It seems to be only a matter of relative yield, and yields do not vary among one another very greatly except for rather easily predictable reasons. Thus with his table of masses the reader can, if

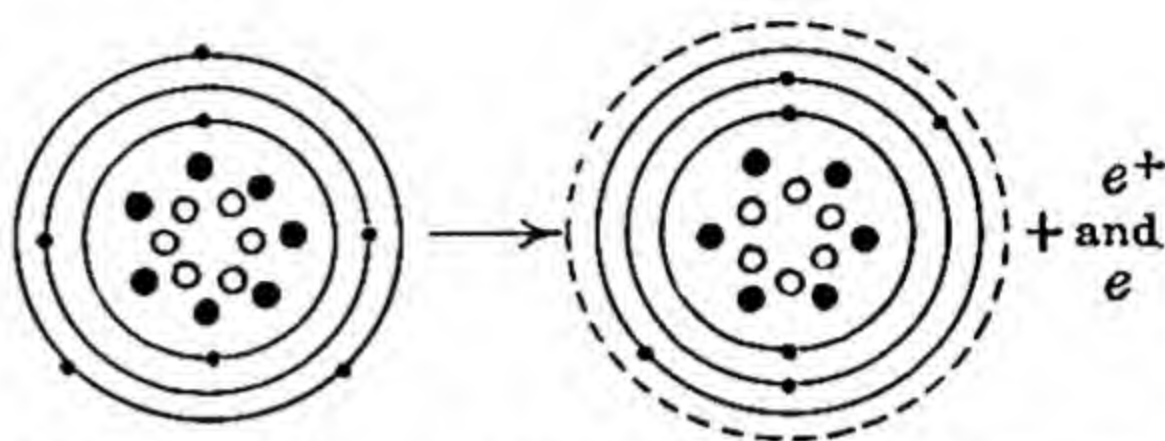


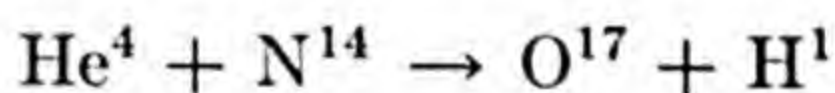
FIG. 2. The transition from radionitrogen,  $\text{N}^{13}$ , to stable carbon,  $\text{C}^{13}$ , shown schematically to illustrate the correction to be made to the masses of the neutral atoms when a reaction in which a positron is emitted is considered. The change itself ejects a fast positron, leaving an atom capable of holding one less electron. Additional mass equal to that of the positron and the electron must therefore be available to make the process go. In the balance sheet this means 0.0011 mass unit on the right-hand side.



he wishes, decide for himself whether a reaction can take place and, if energy will not permit it, discard it. In the second place, since there is a close similarity between types of reactions, a little study of a few categories will be repaid by a reasonably good understanding of the whole subject of nuclear chemistry.

It is a temptation to list here a series of categories of nuclear reactions and then to spend the rest of the chapter discussing them. First, however, we should like to point out that one must keep in mind at once both the nature of the incident particle and the product-ejected particle. For example, one can see that, if a deuteron (mass 2.0147) strikes a nucleus and causes a reaction in which a proton (mass 1.0081) is ejected, there will in general be a release of energy unless the bombarded and product nuclei have a distinctly unfavorable mass difference. We thus expect this type of reaction to be commonly found, and it is. On the other hand if an alpha particle (mass 4.0039) ejects a neutron (mass 1.0089) there will be an absorption of energy unless the bombarded and final nuclei have a favorable mass difference. We therefore expect this type of reaction to be less commonly found unless the alpha particles are very energetic, a conclusion which is nearly true. It is not quite true, as there is, in the lighter elements, a favorable mass difference which helps this type of reaction.

To aid in considering the in-and-out nature of a nuclear reaction a compact notation has been devised as follows. The bombarded element is placed before parentheses, the projectile just inside, the ejected particle next, and finally after the parentheses the product element. Thus Rutherford's pioneer reaction



is written:  $\text{N}^{14}(\alpha p)\text{O}^{17}$ . The  $\alpha$  stands for the alpha particle,  $p$  for the proton; other symbols are  $n$  for neutron,  $d$  for deuteron. This notation may or may not appeal to the reader, but it is convenient, and one soon acquires it as a sort of language. Thus our predictions about deuteron bombardment with emission of protons and alpha-particle bombardment with neutron emission can be said shortly:  $(dp)$  reactions generally are favorable energetically, but  $(\alpha n)$  reactions not so much so. The common forms of reaction are:  $(np)$ ,  $(n\alpha)$ ,  $(pn)$ ,  $(p\alpha)$ ,  $(dp)$ ,  $(dn)$ ,  $(d\alpha)$ ,  $(\alpha p)$ , and  $(\alpha n)$ . Simple or "radiative" capture occurs, and it is written  $(n-)$  or  $(p-)$  or sometimes  $(n\gamma)$  or  $(p\gamma)$ . More abnormal reactions are  $(n, 2n)$  or  $(pd)$ .

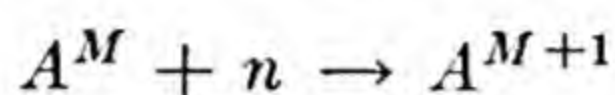
Historically the first class of reaction studied was the  $(\alpha p)$  type. For simplicity we propose to consider neutron reactions first since



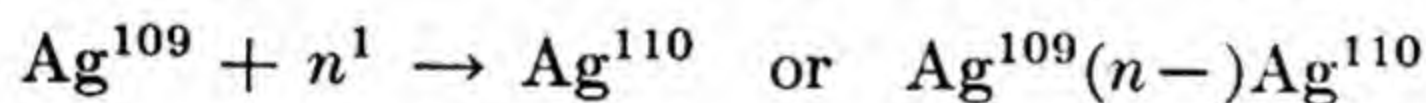
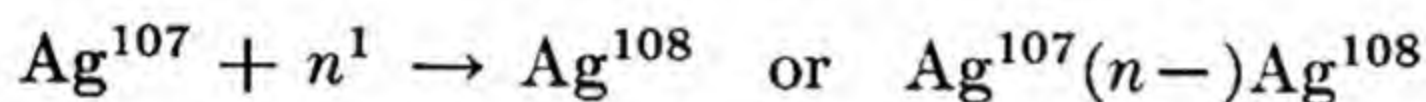
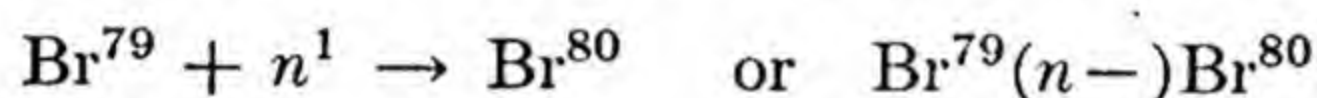
neutrons are free to enter a nucleus without regard to any repulsion set up against their entry. Also, the neutron is so potent an agent in making radioactive materials that it warrants early consideration.

### Neutron-induced transmutations

Only one isotope is known which will not react with neutrons of moderate energy. This is ordinary helium, whose great stability prohibits the creation of any other element from it by combination with a neutron unless very high bombarding energies are used. With this one exception, every element yields to neutron bombardment. By far the commonest process which results from neutron bombardment is *radiative capture*, a process which may be indicated as



meaning that an atom  $A^M$  of atomic weight  $M$  has captured a neutron to form the *composite* and, in this case, also *final* nucleus,  $A^{M+1}$  of weight  $M + 1$ . As illustrations we may take the following, which we have chosen because the reactions can be used for other purposes.



All four reactions result in radioactive products. The first two afford convenient methods of making moderate-sized samples of two of the halogens; the second pair give radioactive silver isotopes which have rather convenient half-lives for use in detecting the presence of neutrons and measuring their intensity. If the reader is ever permitted to approach a cyclotron while it is running and quickly tests the money in his pocket with a Geiger counter, he will find that it is definitely radioactive. The radioactivity will shortly die out and should warn him that the apparently harmless apparatus is nevertheless giving him an appreciable bombardment which will before long begin to have serious effects. The radioactivity will be due to the two last reactions listed above.

This type of reaction is of some interest since it brings up the question of the mechanics of a transmutation. Almost everyone has worked out the familiar problem of two balls colliding and remaining together after collision and knows that it cannot occur if both mo-



momentum and mechanical energy are conserved. Some energy must be lost as heat. Do such considerations hold in the collision of two nuclear particles? If they do, they must clearly have a profound influence on the events in the above type of reaction where the two colliding particles stick together. The answer is yes. The reaction invariably proceeds with the emission of the necessary energy to guarantee the conservation of momentum, not as heat, as in the collision of two putty balls, but as a gamma ray. Now the reader, whether he be a physicist, or a long-suffering physiologist compelled to add one more technique to his already exacting requirements, will know that in atomic physics the emission of radiation is not a continuous process, but that a quantum of energy is emitted as one unit. This fact, that the emitted gamma ray must have a *certain definite energy*, the energy corresponding to the transition between two of a discrete set of levels in the nucleus, means that the outgoing gamma ray, which gets rid of the surplus energy, *can have only certain energy values*. Now the amount of energy to be disposed of depends on the energy of the neutron that does the bombarding. If the amount does not happen to agree with the energy corresponding to a transition in the resulting nucleus, then there is a situation which requires that one of two rigid laws breaks down. In this instance the laws have unquestioned validity, and the only way out is for the process of sticking to fail to take place; to the satisfaction of physicists, this is what happens. The simple capture type of reaction will go for only certain values of the energy of the incident neutrons, those for which the surplus energy exactly fits one of the gamma-ray energies the nucleus is able to emit.

The physicist does not rest content until he has the word "resonance" in every section of his subject, and here it finds its place in nuclear physics. Simple capture is said to be a resonance process: it can occur only if there is resonance between the energy of the incident neutron and the energy of one of the states of excitation of the new composite nucleus. This resonance characteristic has been verified many times. It has some important consequences, because it means that neutrons from any source will not necessarily cause the reaction to proceed and in fact the great variation in the yields of this type of reaction is to be traced to this limitation on the energy of the incident particle.

Let us follow this a little further since it will throw light on our present-day picture of the process of transmutation as well as show what to expect from this particular type of reaction. Modern theory,



for better or worse, has been driven to explain phenomena in the atom by first assigning a "potential field," that is, by deciding the value of the potential energy a particle will have at any point. Reduced to its simplest terms, this means that we cannot say anything about the way a body will move unless we know what forces act upon it. The potential field, which is fundamentally related to the force acting at any place,\* is used simply because it is far more convenient than force to handle in equations; there is nothing inherently abstruse in its use. The necessity of assigning this field in nuclear phenomena is about as cheerless a task as any mathematical physicist has ever had to face, as we are only just beginning to know the forces between single particles, let alone the forces in a complex nucleus. So unhappy a task is it that few theoretical physicists feel optimistic about a complete theory of the nucleus. However, this complexity leads to a certain simplicity, for the theoretical physicist "gives up" and represents the potential field of a nucleus simply by a potential "well" as in Fig. 3.

An incoming neutron is quite unaffected by this well until it is between the points *A* and *B*, where a strong attraction operates on it and it is swept into the complex of particles which comprises the nucleus it is bombarding. The reader must remember that falling into this well is not like the usually imagined accident. The neutron, in falling, acquires speed which may easily carry it up the far side of the well and so out again, without any apparent change at all. On the other hand this entry into the target nucleus is the first requirement of a transmutation and must be considered the preliminary stage. The second stage is concerned with the adaptation of the remaining particles to the new situation. For the reaction under discussion, simple capture, the new composite will be acceptable and it will "settle down" by emitting a gamma ray. We therefore picture the neutron falling into the potential well, acquiring speed which it may

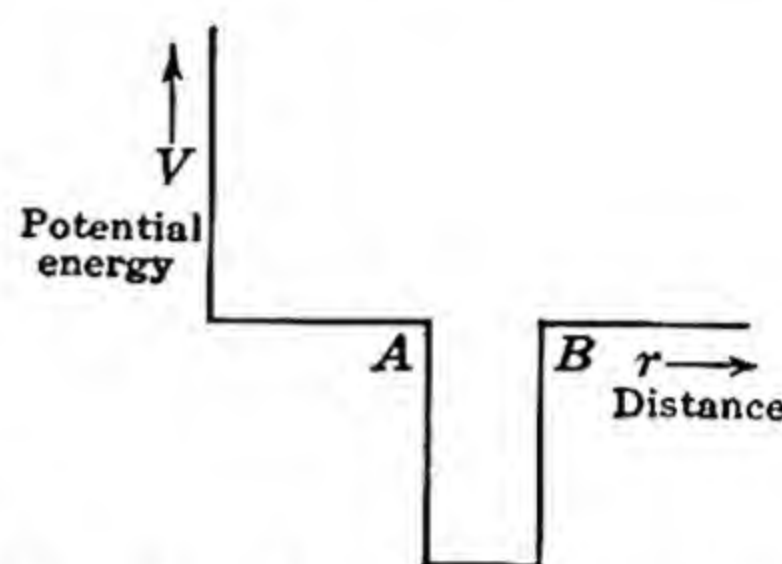


FIG. 3. The potential field of a neutron in the neighborhood of a nucleus, as represented for simple theory. The neutron experiences no force except between *A* and *B*, where very strong forces act. On the potential-energy diagram above, such strong forces are represented by a sudden drop, forming a "potential well," into which the neutron will fall. As it falls it acquires speed and can easily bounce out of the well unless some reason for its "capture" exists.

\* The negative of the derivative of potential energy with respect to distance measured in any direction is the force in that direction.



lose by giving it to the other particles in the nucleus, which bears its abnormal excitation for a short while and then emits a gamma ray to restore itself to a normal condition with the new neutron accepted. Or if the emission of a gamma ray is not feasible, the motion of the particles continues until a neutron has enough energy to duplicate the original energy of the incident neutron, whereupon it is ejected and no transmutation has taken place. The question of the feasibility of emission of a gamma ray turns upon whether in the particular potential well there is a permissible energy value exactly agreeing with the energy the neutron has to contribute. It turns out that all but the lightest nuclei have many available levels, so that the resonance requirement does not cause so great limitation as might be expected. In fact, it can be said quite generally that, with the exception of  $\text{He}^4$ , any element  $A^M$ , after bombardment by neutrons from any but a very peculiar source of exactly one energy, will contain some atoms of  $A^{M+1}$ . Often  $A^{M+1}$  is not detectable as it is stable, and is not produced in sufficient amounts to be detected by ordinary means, but if  $A^{M+1}$  is radioactive, even the small proportion of transmuted material can easily be detected. This generality of neutron capture was discovered in 1933 by Fermi and his collaborators at Rome; it is one of the most important discoveries in nuclear chemistry.

### Slow neutrons

While engaged in this work Fermi found that, if the neutron source were surrounded by water, or paraffin, or any substance containing a large proportion of hydrogen, it frequently was far more efficient in producing radioactivity. Thus if silver is exposed to "straight neutrons" it becomes active. If, however, the silver and the neutron source are enclosed by paraffin about 3 inches thick, the yield is increased about tenfold. It is in fact this combination which is present when a visitor carries money in his pocket near a cyclotron, since the visitor himself is hydrogen-containing material, and the money contains the silver. Fermi was quick to see that the significant fact about the hydrogenous material was the *slowing down* of the neutrons by impact with the hydrogen. It may be recalled that after an impact with a hydrogen nucleus a neutron loses on the average 60 per cent of its energy. This has remarkable consequences after only a few collisions, as is shown in the sequence below.

Number of collisions:	0	1	2	3	6	9	12
Energy (Mev):	5	2	0.8	0.3	0.018	0.001	0.0001



It will be seen that after only a dozen collisions the neutron has only a few electron volts of energy left and that it will require very few further collisions to reduce the speed of the neutrons to that of the molecules of the water or paraffin (speeds corresponding to a few one-hundredths of an electron volt). Such neutrons would be particularly deadly in causing transmutations, for if they happened to strike a nucleus they would spend a long time in its neighborhood with a resulting excellent chance of being caught. This phenomenon of transmutation by *slow neutrons* can often be turned to great advantage and made to yield very concentrated sources of artificially radioactive elements.

The phenomenon of resonance plays an important part in slow neutron reactions, for naturally the majority of the slow neutrons will have energies at or near the energy of the molecular motion, and if it happens that an energy level is not available for this energy the reaction will not be favored. On the other hand if it turns out that an energy level is available there will be a very prolific yield.

This resonance phenomenon has been studied very accurately as will be described shortly. In the meantime a word on the method of description of yield in nuclear processes is in place.

## Cross section

In Chapter 1 we mentioned that nuclear bombardment processes are random in character. Such random processes can only be described essentially in terms of chance. However the chance depends on three factors, all quite obvious. The first is the number of particles which do the bombarding, the second is the number of nuclei in the path of the bombarding particles, and the third is the individual behavior of the nucleus which is being bombarded. The first is readily estimated: if neutrons are responsible there must be some method of measuring their number; if charged particles are responsible the number can be estimated from the current. The second factor can also be estimated if the thickness and composition of the material of the target are known. This estimation can be in terms of Avogadro's number, the number of molecules in a mole, which is  $6.03 \times 10^{23}$ , or in terms of the mass of a hydrogen atom which is  $1.66 \times 10^{-24}$  gram.\*

\* For example, 1 microampere of deuterons is  $6.3 \times 10^{12}$  deuterons per second. A target of aluminum of thickness 16 milligrams per square centimeter (10 cm air equivalent) has  $1.6 \times 10^{-3} / 1.66 \times 10^{-24} \times 27 = 3.57 \times 10^{19}$  atoms of aluminum per square centimeter.



There remains the behavior of the nucleus itself. From the many ways of describing this there has evolved a rather simple and appealing method. This is to assign a target area, or *cross section*, to each nucleus. This target area must be thought of as a synthetic quantity which may bear no resemblance to the physical area of the nucleus, though it probably will often be close to that figure. The unit of cross section,  $10^{-24}$  cm, is often called the "barn." This term was coined during the war by the nuclear physics team at the University of Chicago. It may survive and we hope it will, but scientific linguistics often run counter to usual linguistics because scientists are overly deferent to international conferences.

Now consider how we describe a chance. Suppose that the target contains  $N$  nuclei per cubic centimeter. Suppose the area exposed is  $A$  square centimeters, the thickness  $dx$ . The number of nuclei exposed is then  $NA dx$ . The target area exposed is then  $\sigma NA dx$ , where  $\sigma$  is the cross section per nucleus in square centimeters. The chance of being hit is therefore  $\sigma NA dx/A$ , the target area divided by the whole area over which the bombarding particles are distributed. This is  $\sigma N dx$ , which is interesting because it does not depend on the actual area of the target material.

The yield of transmuted atoms can also be expressed in terms of a chance. Suppose that  $n$  particles bombard the target, and  $dn$  nuclei undergo change as a result of the bombardment. Then the chance can be stated to be the ratio of successes ( $dn$ ) to trials ( $n$ ) and is  $dn/n$ .

Now equating these two expressions for chance (or, more eruditely, probability) we have

$$\frac{dn}{n} = \sigma N dx \quad \checkmark \quad (1)$$

This enables  $\sigma$  to be measured in terms of the yield of the reaction.

It is also possible to consider the way in which the bombarding particles are lost as a result of this process. The particles lost can be written as  $-dn$  so that for *absorption*

$$\frac{-dn}{n} = \sigma N dx \quad \checkmark \quad (2)$$

If integrated this gives

$$\frac{n}{n_0} = e^{-\sigma N x} \quad \checkmark \quad (3)$$



where  $n_0$  is the number of particles incident at the surface of the target. The quantity  $\sigma N$  is often called the *absorption coefficient* for a particle.

Equations 2 and 3 hold for any random absorption process, whether for neutrons, x-rays, photons, or raindrops falling through the leaves of a tree. They are used exceedingly often in nuclear physics.

Cross sections vary from 70,000 barns in some neutron resonances to  $10^{-11}$  barn for electron-induced reactions.

## Neutron resonance capture

The resonance nature of neutron capture was suspected in early experiments. The neatest proof of this was achieved by measuring

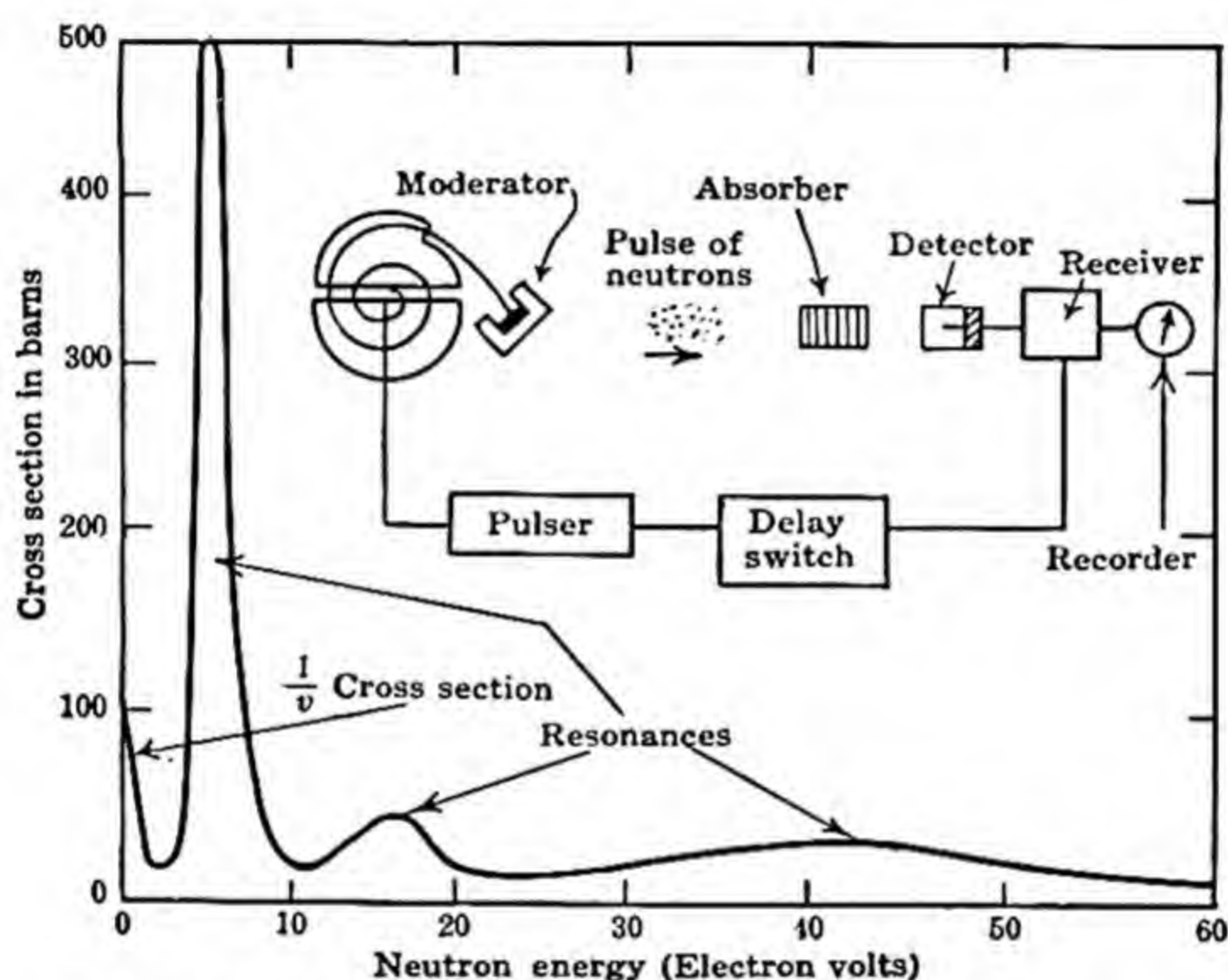


FIG. 4. Cross section of silver as a function of neutron energy. The insert shows the way in which a pulsed beam of neutrons is allowed to travel across a known distance and pass through an absorber. The neutron energy is determined by the time of flight. The total cross section obtained in this way shows a dependence on  $1/v$ , where  $v$  is the neutron velocity, plus three resonance peaks at 5.2, 15.5, and 45 electron volts neutron energy.

the time of flight of neutrons which take part in radiative capture. Early measurements were made with a rotating shutter, but the best method proved to be the modulation of an accelerator. This was first tried by Alvarez, was developed by Manley and Hawthorth and Baker and Bacher, and has been most exploited by the group at Columbia, notably Havens, Rainwater, and Dunning.

Their method is as follows. A cyclotron beam is turned on for a few microseconds by turning on the accelerating voltage on the ion



source. The beam strikes a beryllium target surrounded by paraffin. The burst of neutrons produced by the reaction



is slowed down in the paraffin (an operation which doesn't take very long) and some escape from the paraffin. These are slow and drift relatively leisurely toward an absorber of some selected element. On the other side of the absorber is placed a detector which is turned on only at times which are delayed with respect to the time the ion source is on. These times correspond to various drift times of the neutron and therefore to various neutron speeds. For each such delay time the number of neutrons absorbed is measured by interposing and removing the absorber. Then by equation 3 the cross section can be found. Figure 4 shows the results for silver. The insert shows the arrangement of the apparatus.

The form of the curve is of great interest. For very low energies the cross section is high and smoothly falls off. In this region the cross section is inversely proportional to the neutron velocity (the  $1/v$  region). At the resonance region the cross section obeys the relation

$$\sigma = \frac{\sigma_0 \Gamma^2}{(E - E_0)^2 + \frac{\Gamma^2}{4}} \quad (4)$$

This formula is due to Breit and Wigner and is known as the Breit-Wigner formula.  $\Gamma$  is the "line width." Values which fit Havens and Rainwater data for cadmium, for example, are

$$\sigma_0 = 7200 \text{ barns}$$

$$\Gamma = 0.155 \text{ electron volt}$$

$$E_0 = 0.176 \text{ electron volt}$$

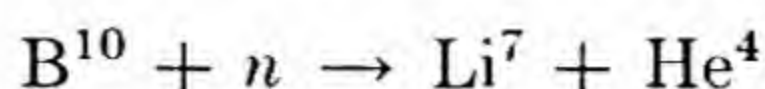
Some resonance cross sections are given in Appendix 10.

The existence of this resonance phenomenon means that there is a great variability of yields in slow neutron reactions so that such means cannot always be applied for the production of radioactive elements.

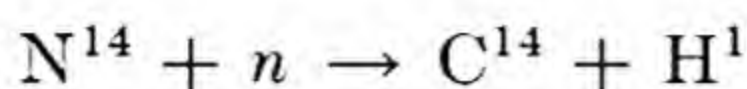
### Neutron-induced reactions with particle emission

Less common than simple capture, but of great importance in some cases, is the phenomenon of *transmutation with particle emission*. Such reactions are

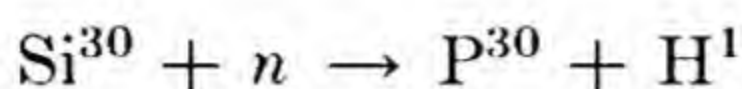




or



or



These reactions have their characteristics fixed for them by the fact that the ejected particle is charged. This has a strange consequence, which at first sight seems absurd, namely, that the charged particle, though it may be given sufficient energy to escape from the composite conglomerate, is still not free to escape. One would naturally conclude that a positive particle would be pushed away all the more easily from a nucleus, and here we are asking the reader to believe that the pushing away is to be considered as a holding in, apparently just at our whim.

This little difficulty is one we are greatly tempted to slide over and beg the reader to accept, but as we are confronted with a property of nuclei which is of the greatest importance we will take a short while to consider it. A few pages back we drew a potential "well" and said that the neutron must fall into this well to start a transmutation. Now let us suppose that it has done so and that a proton is due for ejection. We may now inquire as to the potential well appropriate to this proton and see at once that because the proton is charged it is all higher as drawn in Fig. 5B. Now this shape is clearly wrong, for if the proton were outside the well it would be repelled and thus would run downhill in some potential field; in other words the volcano and crater appearance shown in C is correct. The reader can now see the reason for our paradox. To make a *clean* getaway the proton must somehow acquire enough energy to reach the point *T*. When it does, the repulsion will give it the energy corresponding to a fall down the slope of the volcano so that we would expect our transmutations to yield us only fast particles. This is nearly, but not quite, true. An effect, linked intimately with the property that small-scale matter is governed by the properties of waves, permits the emerging proton to "cheat." A proton of energy *less* than *T* can leak through the walls of the volcano just as light can penetrate a very thin film of gold. This "cheating," or, more elegantly, penetration of the potential barrier, means that in one sense charged-particle transmutations are favored, which is what we at first expected, but that once we have been led to expect this favoring we are to be disappointed in that the process of



leaking through the potential barrier takes time; during that time the nucleus may become impatient and re-eject the neutron instead.

To shorten this account, we find that processes which require charged-particle emission are commonest in light elements where the energy required to get somewhere near  $T$  is small and become less and less common as the nuclear charge increases and the energy of  $T$  becomes greater. To achieve a particle emission from boron a slow neutron will suffice, but exceedingly fast neutrons are needed for elements such as iron. An interesting exception is *uranium*, which is so

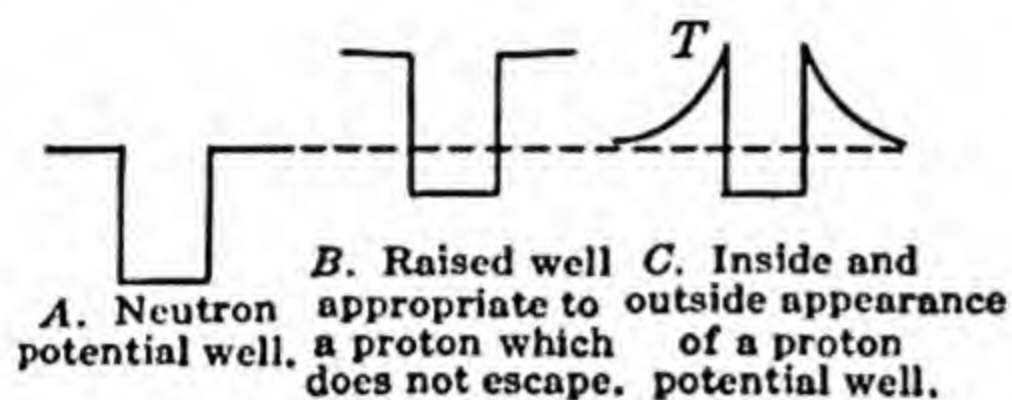


FIG. 5. A diagram to show how the repulsive force exerted on a proton by the protons in a nucleus modifies the appearance of the proton potential well. To be sure of escaping, the proton must have enough kinetic energy to scale the top  $T$ , but it can penetrate the part of the field above zero energy with a certain probability.

need not be resonant since energy and momentum can be conserved for a variety of energies of the emerging particles. The major factor is therefore the opportunity afforded the neutron to enter the boron nucleus. The slower the neutron, the better this is.

Before we go on to consider deuteron-induced transmutations it will be well to summarize the features of neutron reactions. The most common of these is simple capture, which is strongly resonant in nature; the neutron must have one of a set of definite energies or it will be ineffective in producing a transmutation. If one of these energies happens to occur in the region of the energies of thermal agitation, then slow neutrons, or neutrons which have collided so many times that their energy has been reduced to approximately that of the molecular motion, are very effective in producing reactions.

In the next type of process a charged particle is emitted, and here the resonance characteristic is not so important as the fact that the charged particle has to leak out of the potential barrier. The conse-

complex that it is vulnerable to a new type of process in which the whole nucleus can become drawn out into a long droplet shape and then actually blown apart by the mutual repulsion of its charged constituents. Such a process is known as nuclear fission, and a special chapter will be devoted to it.

Where slow neutrons are effective in causing particle emission, most notably in the boron reaction just cited, the cross section depends inversely on the velocity of the neutron. This is because the process



quence is that, unless the neutrons are very energetic, this kind of reaction is not favored, except for light nuclei.

We next consider deuteron-induced transmutations, which are important because the deuteron, having a relatively large positive mass deviation, has a considerable amount of available energy and is potent as a bombarding particle.

### Deuteron-induced reactions

The deuteron is a composite of a proton and a neutron bound together tightly but not inseparably. Its mass is 2.01472, so that it will be expected to be capable of causing reactions in which energy is set free. This is the fact. There are three main types of deuteron-induced reactions: deuteron in, proton out; deuteron in, neutron out; and deuteron in, alpha particle out. In shorthand notation these are  $(dp)$ ,  $(dn)$ , and  $(d\alpha)$ . The general nature of transmutation by deuterons is reasonably easy to predict. The transmutations will not proceed nearly so easily as for neutron reactions, on account of the repulsion exerted by the charge of the nucleus being hit. This repulsion creates a potential barrier which must either be overcome or penetrated (in much the same way as the emergence of a proton from a nucleus is conditional upon either surmounting or penetrating the potential barrier). This greatly limits the number of successful approaches made by a deuteron to a nucleus. Unlike neutrons, which almost all finally succeed in entering a nucleus and causing a transmutation, only about one deuteron in a million manages to do so. It may then be asked why anything but neutrons are ever used for atom smashing; the answer is that neutrons mostly increase the neutron content of a nucleus. Quite often we need to increase the proton content. This can be done by the  $(dn)$  type of reaction or even more so by a  $(d, 2n)$  reaction.

The yield of deuteron reactions is controlled primarily by the number of target nuclei accessible to the deuterons, and this in turn can be analyzed from two points of view. The first concerns the *range* of the deuteron. The great disadvantage of charged particles as transmuting agents lies in the fact that nuclear processes compete disadvantageously with electronic processes, resulting in ionization. This occurrence of ionization stops the deuteron in a definite range so that only those nuclei encountered in this path have a chance of being transmuted. This is the first factor in determining the yield.

The second factor in making target nuclei accessible is the repulsion of the charged deuteron by the charged nucleus. Unless the deuteron



can come within the range of nuclear forces (about  $2 \times 10^{-13}$  cm) it has no chance of producing a reaction. The repellent potential barrier works against this close approach. To see this effect consider the reaction yields plotted in Fig. 6. Note that the yield is zero for zero deuteron energy (in contrast to neutron reactions) and rises in each case in a more or less exponential way at first. The heavier the element the higher the energy before the yield is appreciable. After the

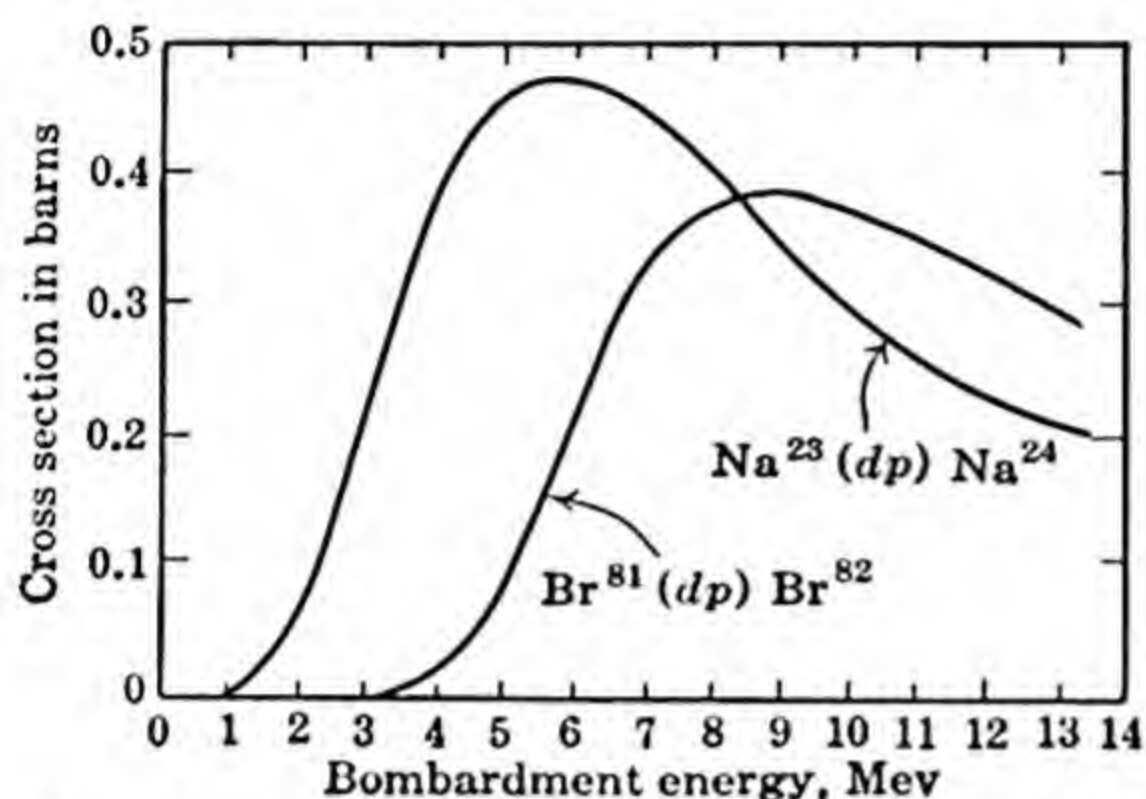


FIG. 6. Yield curves for deuteron reactions taken from data of Clarke and Irvine. The initial rise is due to penetration of the barrier; the falling off is probably due to competition by other processes. The barrier is higher for Br than for Na because the nuclear charge is greater.

yield has risen considerably there is a tendency to stabilize or even fall off.

The initial rising part is due to penetration of the potential barrier by the deuterons. The flattening is due to the fact that the bombarding deuterons have sufficient energy to go over the top of the barrier. Any fall in yield at these high energies is most likely to be due to the diversion of deuteron action to other processes not detected by the method employed. Thus if the emission of neutrons is being observed and a second process involving the emission of protons becomes pro-

liferous, this will detract from the neutron yield. This feature of the *competition between various processes* is most important in nuclear reactions.

Some rough rules for estimating the effect of the potential barrier can be given. In the first place it has been found that the radii of nuclei are governed by the simple proportionality between *the volume of the nucleus and the number of particles in it*. This gives the actual expression

$$r_0 = 1.5 \times 10^{-13} \times A^{1/3} \text{ cm} \quad (5)$$

for the radius  $r_0$  of a nucleus of atomic weight  $A$ .

The cross section can then be estimated to a first approximation as  $\pi r_0^2$  or  $7.1 \times 10^{-26} A^{2/3}$  sq cm. This can be regarded as par for the cross section. As we have already seen, quite wide variations are possible.

The fact that deuterons must surmount or penetrate the barrier before they have a chance to act on this cross section means that the height



of the barrier is important. The energy of the top of the barrier is equal to

$$\frac{Ze^2}{r_0} \text{ ergs} \quad (6)$$

where  $Z$  is the nuclear charge and  $e$  is the charge on an electron in electrostatic units. (Note that the energy is doubled if an alpha particle, doubly charged, approaches.)

For the element carbon,  $A$  is 12 so that  $A^{1/3}$  is 2.29 and  $r_0$  is  $3.44 \times 10^{-13}$  cm. Since  $Z$  is 6, the energy of the top of the barrier  $E_T$  is

$$\frac{6 \times 4.8^2 \times 10^{-20}}{3.44 \times 10^{-13}}$$

or  $4.0 \times 10^{-6}$  erg. This is 2.5 Mev.

Equations 5 and 6 can be combined to give the approximate formula for singly charged bombarding particles,

$$E_T = 0.9 \frac{Z}{A^{1/3}} \quad (7)$$

where  $E_T$  is in million electron volts.

Knowledge of the value of  $E_T$  at once permits a rough estimate of a nuclear yield. For values of the bombarding energy in excess of  $E_T$ , the cross section will be about  $7.1 \times 10^{-26} A^{2/3}$  sq cm. For  $A = 125$  the yield for a target containing  $10^{19}$  atoms per square centimeter would be  $177 \times 10^{-26} \times 10^{19}$  or approximately  $1.8 \times 10^{-5}$ . This means one transmutation for every 55,000 deuterons. Experiment roughly verifies this.

A better estimate which takes into account barrier penetration can be made as follows. The probability of penetration through a barrier is given by the relation

$$P = e^{-4\pi/h \int_{r_0}^{r_1} 2m(V-E)^{1/2} dr}$$

where  $h$  is Planck's constant,  $r_0$  and  $r_1$  are the inside and outside radii of the barrier,  $m$  is the mass of the bombarding particle,  $V$  is the potential energy of the bombarding particle at distance  $r$ , and  $E$  is the bombarding energy. All energies are in ergs.

The value of  $r_1$  is given by the relation

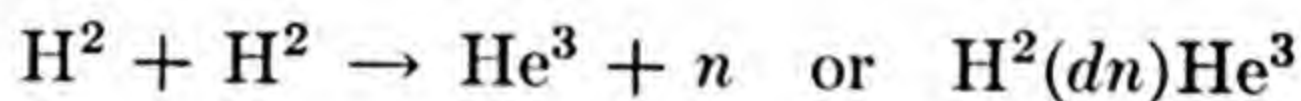
$$E = \frac{Ze^2}{r_1}$$



Now a glance at the exponent in this equation shows that when the integral is zero the probability is unity. In order for the probability to diminish to 37 per cent the integral must be equal to  $h/4\pi$ , for then the whole exponent is  $-1$  and  $e^{-1}$  is about 0.37. Simple trial shows that this figure is reached for  $E$  about 60 per cent of  $E_T$ . Hence, penetration of the barrier is quite efficient near the top. As  $E$  diminishes to  $E_T/10$  the probability rapidly diminishes.

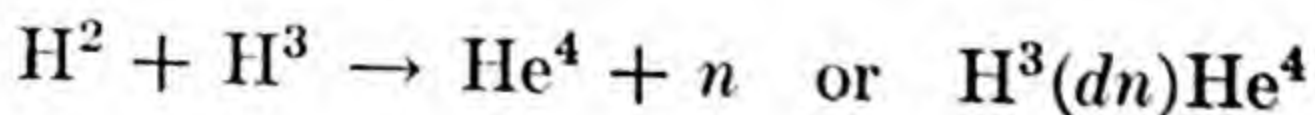
To find the actual yield in a reaction it is necessary to estimate the target thickness in terms of the penetrating ability of the deuteron. This whole thickness is then divided into manageable sections, and the cross section for each is figured. The cross section multiplied by the number of nuclei exposed in each section gives the overall cross section per section. Adding these gives the chance a single deuteron has of causing a reaction. The reciprocal of this is the number of deuterons needed to produce a single transmutation.

Having gone over the generalities of this type of reaction, let us consider a few important examples. The first is a series of reactions which yield neutrons. The simplest is the bombardment of deuterium by deuterons. The reaction is



This reaction is of great importance because it involves nuclei of very low charge and therefore proceeds at extremely small energies of the bombarding deuterons. The yield was so great when the original discovery was made by Oliphant, Harteck, and Rutherford in 1934 that it was thought to be due to extraneous x-rays until check experiments showed the actual nature of the process. There is no accompanying gamma radiation, a fact which simplifies the interpretation of experiments made with neutrons from this as a source. If an inexpensive installation of a source of neutrons is needed, a moderate yield can be obtained by adapting a 100-kilovolt transformer to accelerate deuterons which then bombard a target of heavy ice. Sufficient numbers can be obtained to prepare fair samples of several elements for tracer work, for example, chlorine, bromine, and iodine. It has proved possible to detect the presence of neutrons when the bombarding energies are less than 10,000 electron volts.

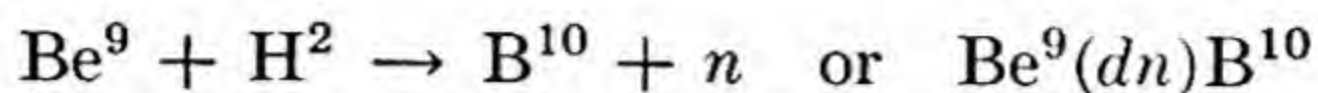
Similar in character to this is the bombardment of tritium by deuterons. The reaction is





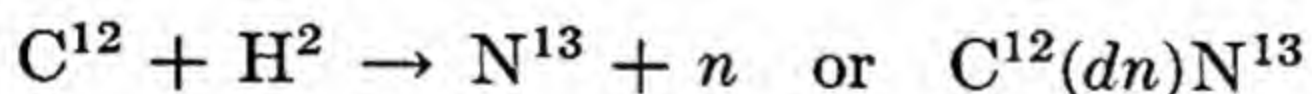
The yield is very large, and this reaction is a convenient source of high-energy neutrons of one energy.

The next two reactions to be considered are also important as neutron sources. The most commonly used neutron source is the reaction



At bombarding energies above 1 Mev the yield of neutrons from this reaction exceeds that from any other of this type, and as beryllium is very stable to heat it forms an ideal target.\* It is likely that beryllium is the most-bombarded element, almost solely on account of its use in this way. The neutrons so produced are, with cyclotron deuterons of 5-Mev energy, spread out between 0 and 9 Mev. Such neutrons are fast but not the fastest that can be obtained, and they will not cause all the reactions which can be induced by neutrons, notably the type in which for the entry of one neutron two are ejected, the  $(n, 2n)$  type. The fastest available neutrons are produced by bombarding lithium by deuterons; here the energy change is roughly 15 Mev, so that with 5-Mev deuterons the energy produced is nearly 20 Mev. The yield is also large, not much less than from beryllium, and so this form of neutron source finds considerable application also. The difficulty is to find a stable target which will withstand long hours of bombardment and yet contains a high proportion of lithium. If the target is well cooled, lithium itself can be used. Lithium nitride is a very stable compound.

A fourth reaction of this type should also be mentioned since it accompanies virtually every deuteron bombardment in spite of all precautions. This is the reaction

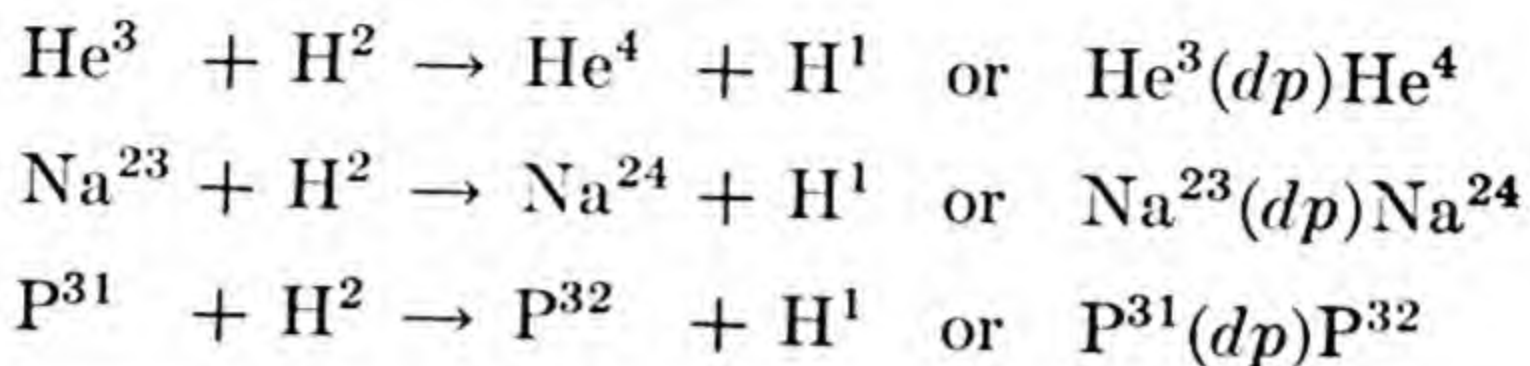


The presence of carbon as impurity in any material other than that which has been treated with extraordinary precautions is almost universal. It is therefore found that after removing a target from a bombardment there is always a 10-minute positron activity due to the decay of the radionitrogen formed. This activity must be considered in all experiments in which the time of working occurs soon after the bombardment. It is easily avoided, if time is available, by letting the product "age" for an hour or so, after which the amount of radionitrogen present is negligible.

\*Beryllium dust is highly dangerous; the handling of beryllium should be treated as an industrial hazard and care taken accordingly.



A second deuteron reaction of interest is that in which a proton is ejected, in shorthand the  $dp$  reaction. The reader will notice that, if a deuteron enters a nucleus and a proton is ejected, the whole is the equivalent of the addition of a neutron alone. Therefore the  $dp$  type of reaction achieves the same result as simple capture in the bombardment by neutrons. Examples are

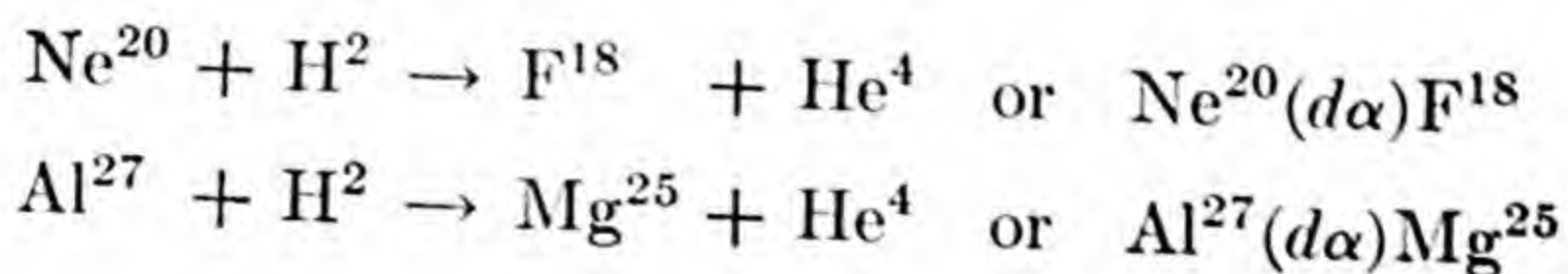


Of these the second and third have been used very extensively to produce radiosodium and radiophosphorus.

The above ( $dp$ ) type of reaction is of interest for the particular reason that it involves a unique method for the process of the reaction. The deuteron does not have to penetrate all the way into the nucleus to cause a transmutation; it can cause one by merely getting near the repulsive field of the bombarded atom. In the repulsive field the deuteron ceases to act as a single particle. It is a composite of a neutron and a proton, only one of which experiences any effect due to the charged nature of the target nucleus. The proton is repelled away from the nucleus, while the neutron is not affected, and the result is a tendency for the neutron and the proton to separate. If this happens, the neutron is conveniently near the target nucleus and is captured by it, while the proton is sent away. The resemblance to the simple capture of a neutron is thus not merely trivial; in a real sense the process is actually such a capture, only the neutron has to be specially created from a deuteron in the neighborhood of the nucleus. This method of effecting a reaction was suggested by Oppenheimer and Phillips, and the  $dp$  type of reaction is often referred to as the Oppenheimer-Phillips reaction.

At bombardment energies above about 10 Mev the process of "stripping" can take place. The target nucleus captures a neutron or a proton from the deuteron, and the residual particle flies on alone. The yield in this process is large and is predominantly in the same direction as the incident deuteron.

A third type of reaction has already been mentioned: the  $d\alpha$  type. As examples of this we have





The first gives a 110-minute half-life isotope of fluorine; the second gives a stable form of magnesium. This type of reaction is not of great use in preparing radioactive materials as the yield is not so great in general, owing to the higher barrier to the emergence of the doubly charged alpha particle. Nevertheless it should be considered a possible means of forming a desired radioelement.

Under the head of deuteron reactions we have considered many of the features common to reactions involving charged particles. Therefore we can consider the remaining two groups of such reactions rather more briefly. These two groups are proton- and alpha-particle-induced reactions. Since alpha particles can be accelerated in a cyclotron set up for deuterons without any change, it is not bad to group alpha-particle reactions with deuteron reactions.

### Alpha-particle reactions

The main feature of interest about alpha-particle reactions is that they produce a considerable change in the mass of the bombarded element. That is to say, one produces an element 3 mass units away from the original element. This may be a useful method of reaching an isotope required for certain purposes.

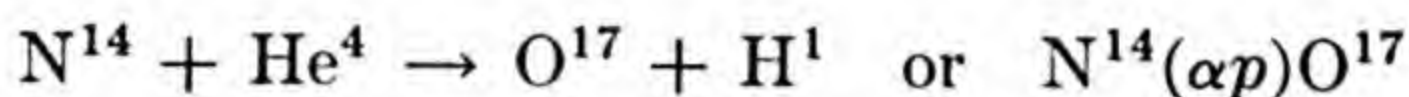
A second feature of alpha-particle reactions is the small mass excess of the helium atom, which it will be recalled means that relatively little mass energy is available to make the reaction proceed. This is largely compensated for by the fact that the double charge of the alpha particle enables the same cyclotron to accelerate helium nuclei to double the energy of the corresponding deuterons. To take a definite example we may consider the beams available from a 60-inch cyclotron. The deuteron beam is 20 Mev; the alpha-particle beam is 40 Mev. To the deuteron energy must be added the mass excess of 13 Mev equivalent; to the alpha-particle beam, the smaller mass excess of 4 Mev equivalent. The deuteron thus totals 33 Mev, whereas the alpha particle is worth 44 Mev and is thus already more able to supply energy needed to make a reaction go than the deuterons produced by the same cyclotron. This situation is not true for low-energy beams, where the deuteron is an easy winner, but it can be seen that the effectiveness of alpha-particle bombardment by a large atom smasher must not be underestimated.

### $\alpha p$ and $\alpha n$ reactions

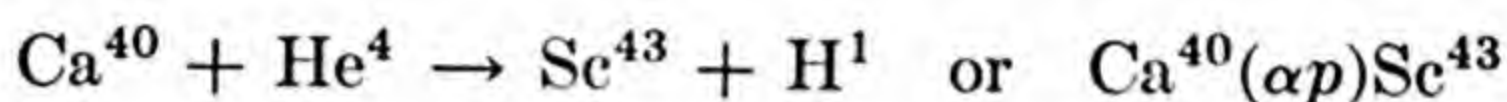
In considering the  $\alpha p$  type of reaction, although we may give several historically interesting reactions as examples of this kind of



transmutation it should be pointed out that it is of little importance in the manufacture of artificially radioactive substances. The reason is that the very nature of the reaction requires the addition of two neutrons and one proton to the bombarded nucleus, and, as the number of neutrons relative to protons is gradually increasing as we proceed towards elements of larger mass, the reaction will tend to produce stable nuclei, at least in the lighter elements. In the heavier elements this is not so true, although it still remains a type of reaction which is suitable for studying transitions between stable nuclei rather than a reaction for producing radioactivity. As examples we may choose



This is the pioneer reaction of Rutherford which has ushered in the whole subject of modern nuclear physics. The  $\text{O}^{17}$  is stable.



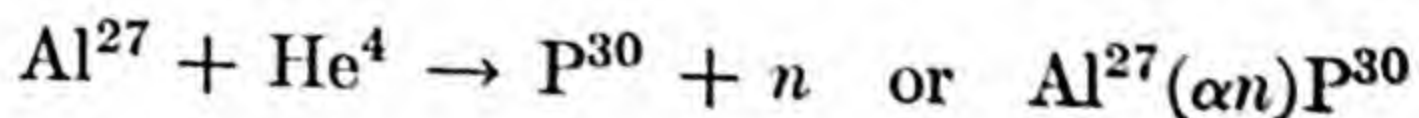
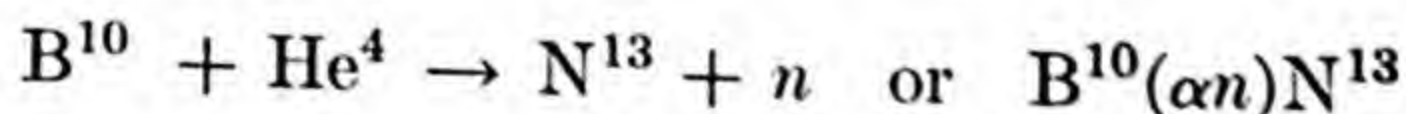
This reaction results in the formation of a radioactive form of scandium having a half-life of 4 hours.

The  $\alpha n$  type of process in which a neutron is emitted is of more practical interest. It is of some importance as the first method by which neutrons were produced, and indeed the reaction



is still commonly used as a neutron source when either no better is available or for some experimental reason the whole space surrounding the source must be filled with some material under study. Such a source commonly consists of powdered beryllium mixed with radium. A reasonable estimate of the number of neutrons emitted by a mixture of 1 millicurie of radium and beryllium is 25,000 per second. This can easily be exceeded by almost any equipment using artificial acceleration, but it is sufficient for many purposes.

The  $\alpha n$  reaction as opposed to the  $\alpha p$  type increases the ratio of protons to neutrons and so tends to cause reactions which result in unstable nuclei. Historically the discovery of artificial radioactivity was made by means of this reaction. We may cite the reactions first discovered by Curie and Joliot.





In both these reactions the final nucleus has too much charge and relieves itself of the excess by the emission of a positron. This type of reaction generally produces positron emitters. The  $N^{13}$  is already familiar from the bombardment of carbon by deuterons and decays with a 10-minute half-life.  $P^{30}$  decays with a half-life of 2.5 minutes. Finally we come to the use of protons as the bombarding particle.

### Proton-induced reactions

The proton was the logical particle to try in the first experiments with artificially accelerated particles. It was found to be eminently successful, as the reader knows. The pioneer work of Cockcroft and Walton, shortly followed by many others, showed that by bombarding light elements with protons a variety of reactions could be made to go. Since we are primarily interested in presenting a systematization of nuclear chemistry we do not intend to consider the historically interesting reactions but will follow the same order as for the neutron reactions and will start with *simple capture*.

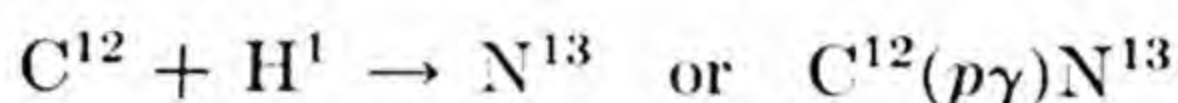
The astute reader may perhaps wonder why we made such a business of simple capture for neutron bombardment, ignored it for deuteron and alpha-particle bombardment, and now return to it for proton reactions. The reason is mainly experimental. There is now some evidence for simple capture of deuterons, but in general the many products involving particle emission obscure the process of simple capture, which has to be detected by observing the emitted gamma radiation that it causes. Also the great mass excess of the deuteron means that, when it is captured, the resulting conglomerate is in a high state of excitation and in general seems to prefer to emit a particle rather than settle down by mere gamma-ray emission. This, of course, only tells us that the process is uncommon, not that it does not occur. It does tell us that it will be hard to detect. Similar experimental conditions also prevent the detection of this process in alpha-particle reactions. For both neutrons and protons the bombarding particle is elementary, and this greatly aids the detection of the simple capture process. It means that the whole reaction is simpler and so the competition from other processes is less.

The results of the simple capture of protons are of the greatest value in many research experiments. The gamma ray which is evolved after the proton has been caught is likely to be very energetic; for example, the bombardment of lithium yields a gamma ray of energy 17 Mev, far greater than the 2.6-Mev ray which is the most energetic available



from natural sources. Other reactions give less energy than this, but the run of energies is around 6 Mev, which is still considerable. Since the yield of such gamma rays is great and beams of protons of several microamperes can be used, these reactions can be made to act as sources of energetic gamma rays which can, themselves, produce transmutations. One does not use gamma rays for actual manufacture of radioactive materials, but they have been of great value in nuclear research.

In addition to the emission of an energetic gamma ray, the capture type of reaction can give rise to considerable yields of radioactive materials. To cite a simple example, the bombardment of carbon by protons causes the formation of a substance with the familiar 10-minute half-life associated with  $N^{13}$ . The reaction is



The radioactivities associated with the radiative capture of protons are the same as would be produced by the  $dn$  type of reaction on the same target. If a cyclotron is set to be in resonance for protons it can still be used to give many radioactivities produced by deuteron bombardment.

The salient feature of simple capture of neutrons was shown to be resonance. Since there is no essential difference between proton bombardment and neutron bombardment as far as the formation of a composite nucleus is concerned, it is to be expected that there will be resonance in proton bombardment. This is so. If a suitable gamma-ray detector is placed near a target of lithium bombarded by protons whose energy can be smoothly varied (as, for example, protons from a Van de Graaff generator), there is found to be no yield of gamma rays at all until the energy of the beam exceeds 440 kilovolts. At this energy the yield of gamma rays suddenly shows a marked increase, and, if the layer of lithium is thin so that it does not cause any slowing up of the protons, the yield of gamma rays falls as soon as this energy has been passed. At higher energies other resonance levels may become apparent. In simple capture with aluminum as the target, there have been shown to be thirty or more resonance levels between zero and 2.6 Mev. This rapid increase in the number of levels is expected on the present theory of nuclear structure.

After the simple capture process, the next of interest is the  $pn$  process in which the proton is merely exchanged for a neutron. This type of reaction has two characteristic features. The first is that the energy of the proton must always exceed a certain threshold value be-



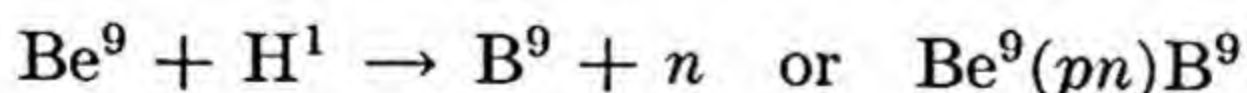
fore the reaction will proceed. The reason for this is that the neutron is heavier than the proton and also that we are always starting with a stable nucleus in the target and producing a less-stable nucleus, which is an abnormally heavy nucleus. This twofold demand on mass requires that we supply the equivalent energy in the form of the kinetic energy of the incident particle; and also that there can be a threshold energy below which we have not supplied enough to make the reaction go. The second characteristic feature is that a positron-emitting radioactive element universally results. It is a rule, which is nearly perfectly obeyed, that no two nuclei of the same mass and charge differing by only one unit can exist. In other terms, if there are two neighboring isobars, one always decays into the other. Isobars which are stable exist, but their charges generally differ by two units. Now if we put a proton into a stable element and take a neutron out, we produce an isotope which has the same mass but a charge differing by one unit from the original element. The new isotope is therefore unstable, and to return to stability it must lose charge, or emit a positron. So a laboratory which specializes in this reaction gets to regard the positron as a commonplace.

As an example of this type of reaction we may cite the formation of  $F^{18}$  from oxygen ( $O^{18}$ ).



This is interesting as it indicates that the yield of this type of reaction is considerable. The amount of  $O^{18}$  present in ordinary oxygen is less than 1 per cent, and yet the yield of radioactive material is readily detectable with insensitive apparatus.

Since it is a handicap to be unable to make any installation deliver neutrons at a moment's notice, it is important to find a reaction which will give neutrons when protons are the bombarding agent. The  $pn$  reaction is suitable, and it is found that if beryllium is bombarded by energetic protons the numbers of neutrons are very nearly as great as if deuterons were used as projectiles. The reaction is

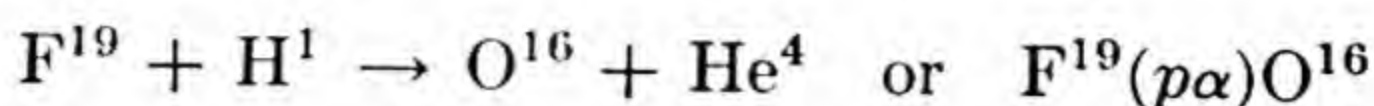


The  $B^9$  is, as expected, a positron emitter.

The last important reaction type is the  $p\alpha$ . There is no striking feature of this reaction, which is the reverse of the  $\alpha p$  type. Except for the lighter nuclei, where this reaction often goes with considerable release of energy, there has been no direct study of the emitted alpha particles, and so the occurrence of the reaction is inferred from the



radioactivity resulting. It is a reaction which is highly favored only at high bombarding energies as the ingoing and outgoing particles are both charged and subject to the necessity of passing through barriers. As an example we may take



### Scattering

By now the reader will almost certainly be wondering whether it is absolutely necessary that a bombarding particle enter a nucleus and combine with it. The answer is no. In a considerable number of cases the particle is merely deflected by the nucleus with which it collides, and when this occurs the process is spoken of as *scattering*. The biggest agent in scattering is, of course, the extensive coulomb field, and the nature of this process was determined by Rutherford, Geiger, and Marsden. The cross section  $d\sigma$  for scattering through an angle  $\theta$  for a light singly charged particle by a heavy nucleus is

$$d\sigma = \frac{e^4 Z^2 \cdot 2\pi \sin \theta d\theta}{16E^2 \sin^4 (\theta/2)} \quad (8)$$

Here the symbols have the usual meaning. The term  $2\pi \sin \theta d\theta$  simply expresses the solid angle subtended at angle  $\theta$  by an angular increment  $d\theta$ . For a doubly charged scattered particle  $d\sigma$  is four times as great.

Such scattering is referred to as *Rutherford scattering* and really has nothing to do with nuclear processes. When a charged particle approaches a nucleus it enters this rising terrain of coulomb repulsion and moves accordingly. If, however, it comes within the grasp of nuclear forces this is changed. A new type of scattering, called *anomalous scattering*, is produced.

It is a pity that spherical geometry involves rather nasty mathematics, for this fact alone prevents us from giving a more complete account of the scattering process. What occurs is a form of interference. The incident particle, which as the reader knows must sometimes be treated as having wave properties, can be thought of as a plane wave. The coulomb field distorts this, but in a definite way. The nuclear field, however, distorts a fraction of the plane wave so as to cause a phase shift. This part of the wave then interferes with the undistorted part and as is well known in interference phenomena produces maxima and minima.



This is beautifully shown in some experiments of Bender, Shoemaker, Kaufmann, and Bouricius. The arrangement is shown schematically in Fig. 7a. Protons of carefully controlled energy are incident on a thin layer of aluminum. Behind this are placed a series of Geiger counters to detect gamma radiation, and at the same time scattered protons which emerge at about 135 degrees to the original detection are collected and recorded by a proportional counter. The result of the gamma ray detection is shown in Fig. 7b. It can be seen

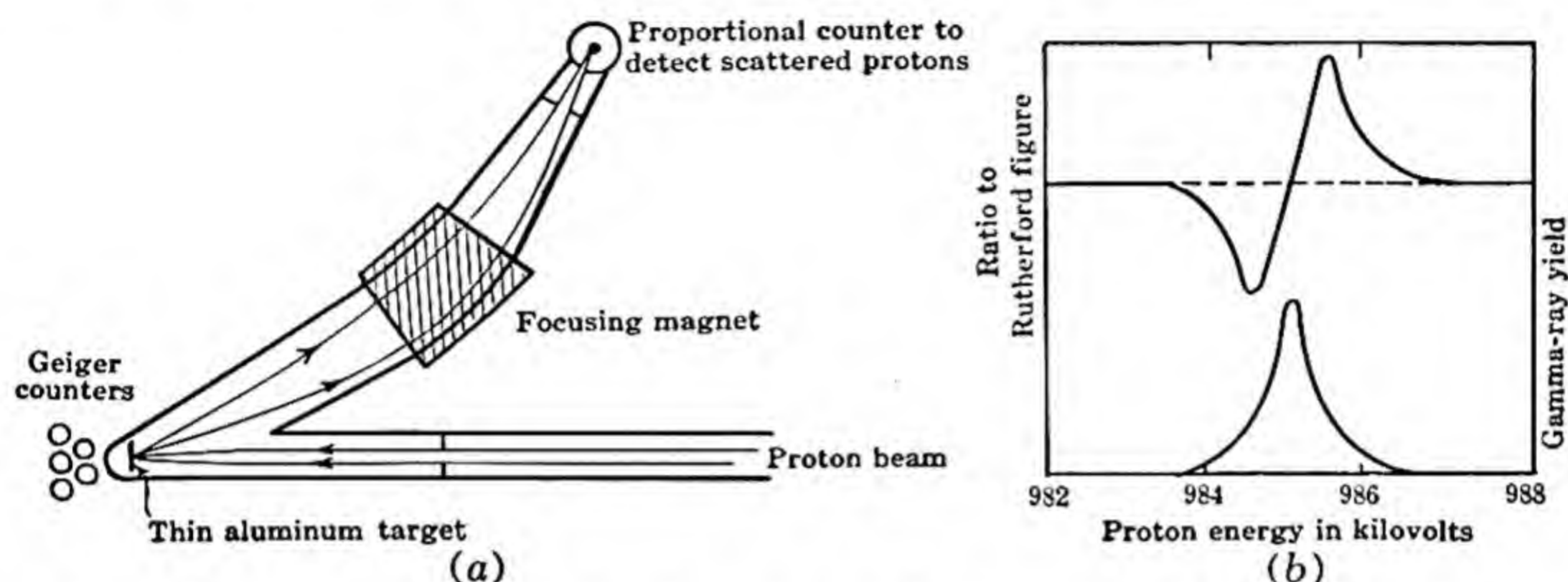


FIG. 7. (a) The arrangement for detection of both scattered protons and gamma radiation when a thin target of aluminum is bombarded by protons of variable energy. The protons scattered backwards are focused and counted in a proportional counter. Gamma rays are detected in five Geiger counters placed behind the target. (b) The yield of gamma radiation as the proton energy increases, and in the upper curve the scattered protons plotted in terms of the ratio of observed to Rutherford theory. The effect of a nuclear energy level on the scattering is clearly seen.

that the cross section for the formation of gamma radiation rises sharply at 985 Kev and falls again. There is therefore resonance with a level at that energy, and this means for sure that protons must be capable of penetrating the nuclear field at that energy. Such protons must therefore be anomalously scattered, and that they are is shown in the upper curve of Fig. 7b. The ratio of scattered protons to the number expected from the Rutherford theory falls for proton energies just below resonance and rises thereafter, falling when the proton has passed completely by resonance.

A further variety of scattering is in reality closer to a transmutation. This is *inelastic scattering*. Here the proton, or other particle, which penetrates the nuclear field is absorbed but emerges with lower energy. The energy lost is given to the nucleus which later parts with this as gamma radiation. As will be shown later, inelastic scattering provides considerable information about nuclear energy levels.



### Process of a transmutation: energy of emitted particles

We have described several kinds of reactions and there now arises the question of how they actually happen. Perhaps we should rapidly state that we do not really know, but this is how we think they happen.

The incoming particle is supposed to combine with the target to form what is called the compound nucleus. This compound nucleus has energy levels, like an atom has energy levels, and if the incoming

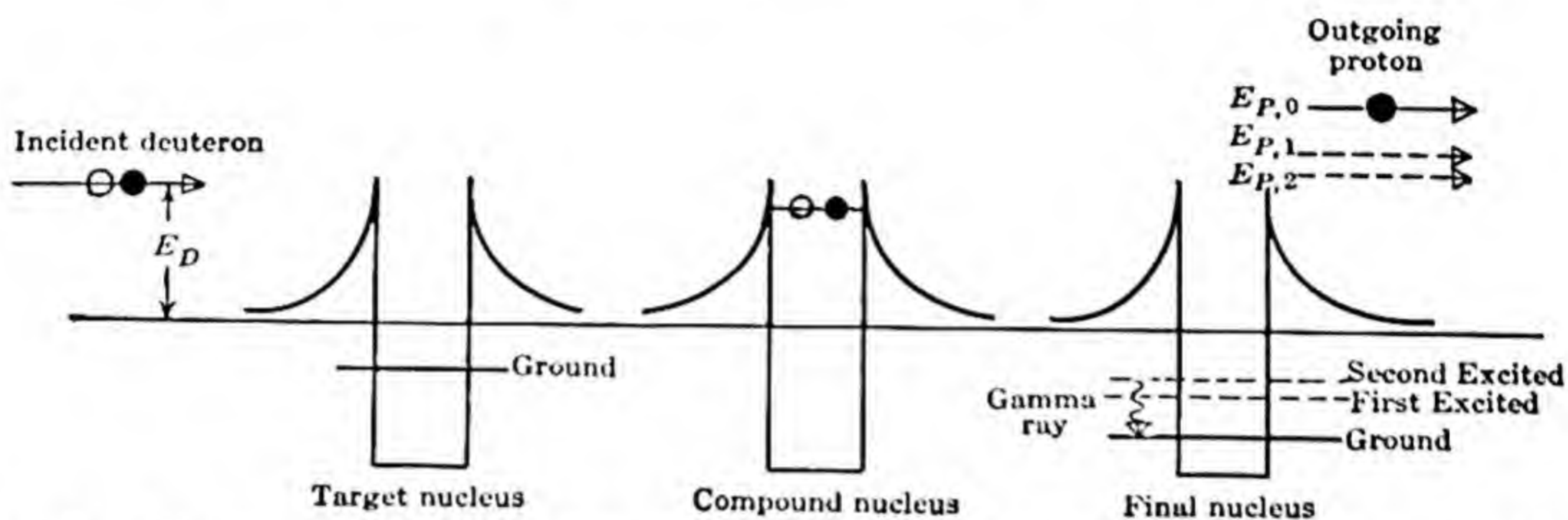


FIG. 8. Illustrating the process of a low-energy transmutation. The incident particle (here a deuteron) enters the target nucleus either through or over the barrier and forms a compound nucleus in a very excited state. This then settles down in various ways, the one illustrated being by emission of a proton. If the final nucleus is left in an excited state the proton has less than the maximum energy. The energy difference between the ground state of the target nucleus and the energy level of the final nucleus is the nuclear energy change, or  $Q$  value.

particle has an energy which is correctly matched to one of these the capture of the bombarding particle is more likely. This accounts for the resonance in radiative capture where the compound nucleus is also the final product. It may well happen that there is an excess of energy levels available, or that the levels are very wide. If this is so the resonance in the formation of the compound nucleus may not be observed as capture can occur over a spread of energy values. This is the case for deuteron reactions where the high excess mass of the deuteron guarantees that the compound nucleus is highly excited.

The compound nucleus is usually very short lived, especially if it is highly excited. It either settles down by radiation (radiative capture) or it emits one or more particles or both, and this process takes place in something less than  $10^{-15}$  second. Now this time is so short that the compound nucleus never collides with anything before it makes its adjustment and therefore it has no method of losing momentum except to the products of the reaction.



This fact that momentum is conserved enables deductions to be made regarding the energy change in the reaction. It will be recalled that this is determined by the mass difference of the reacting particles and the reaction products. An equation relating the nuclear energy change  $Q$  and the various energies involved is given in Appendix 7.

One early result, established by Bothe and Franz and by Chadwick, Constable, and one of the authors, showed that the emerging particles do not have a general spread of energies but have certain definite energies associated with definite  $Q$  values.

This can be understood by looking at Fig. 8 where the various energy relations are shown. The left-hand figure shows a deuteron of energy  $E_D$  approaching a target nucleus represented by the potential barrier and well, as shown. After the deuteron enters the nucleus the compound nucleus is formed as in the center figure.\* This reverts to the final nucleus, for example, by emitting a proton. If this nucleus is formed in the ground state the outgoing proton has all the mass energy available and leaves with energy  $E_{P,0}$ , as shown. However, the nucleus can be formed in an excited state, which withholds some of the available mass energy. The proton then leaves with energy  $E_{P,1}$ , and the difference in energy between  $E_{P,1}$  and  $E_{P,0}$  is therefore the withheld energy or *the energy of the first excited state of the final nucleus*. If the final nucleus is in a higher excited state the proton has less energy. Thus the proton energies represent a kind of inverted picture of the energy levels of the final nucleus.

If it happens that the same process occurs with emission of the incident particle the same kind of information results. This is what oc-

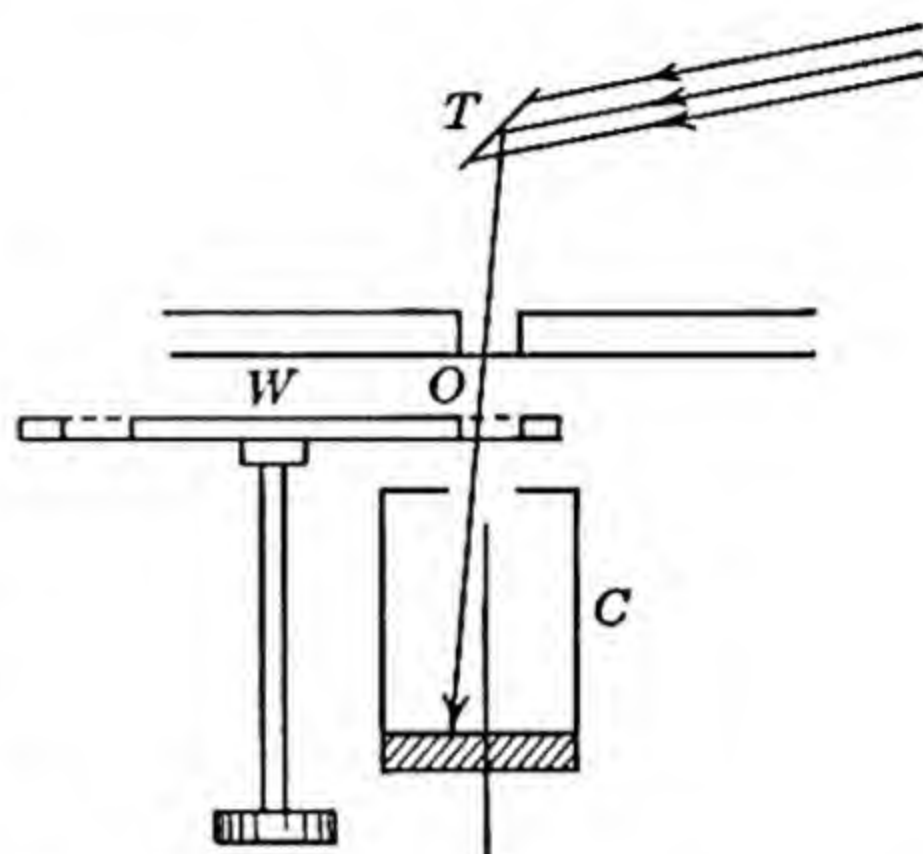


FIG. 9. Schematic arrangement for plotting an absorption curve for protons. A particle beam strikes a target  $T$ , causes the emission of protons, some of which pass through the defining opening  $O$ , and the absorbing foil into the counter  $C$ . The wheel  $W$  carries foils of different thicknesses which can be interposed in front of the counter.

\*The physical chemist will readily see that the compound nucleus plays the same part as the "activated state" in reaction kinetics. The major difference lies in the fact that adequate bombarding, or mass, energy is supposed to be available for the formation of the compound state, whereas in reaction kinetics the critical complex plays a part comparable to a barrier.



curs in the process of inelastic scattering and explains why inelastic scattering is important in energy level research.

In Fig. 9 we show an experimental arrangement for bombardment and product particle detection. The product particles, if they are protons or alpha particles, can be studied by absorption or magnetic analysis. Absorption measurements employ the range-energy relationships given in Appendix 7. It is usual to take advantage of the

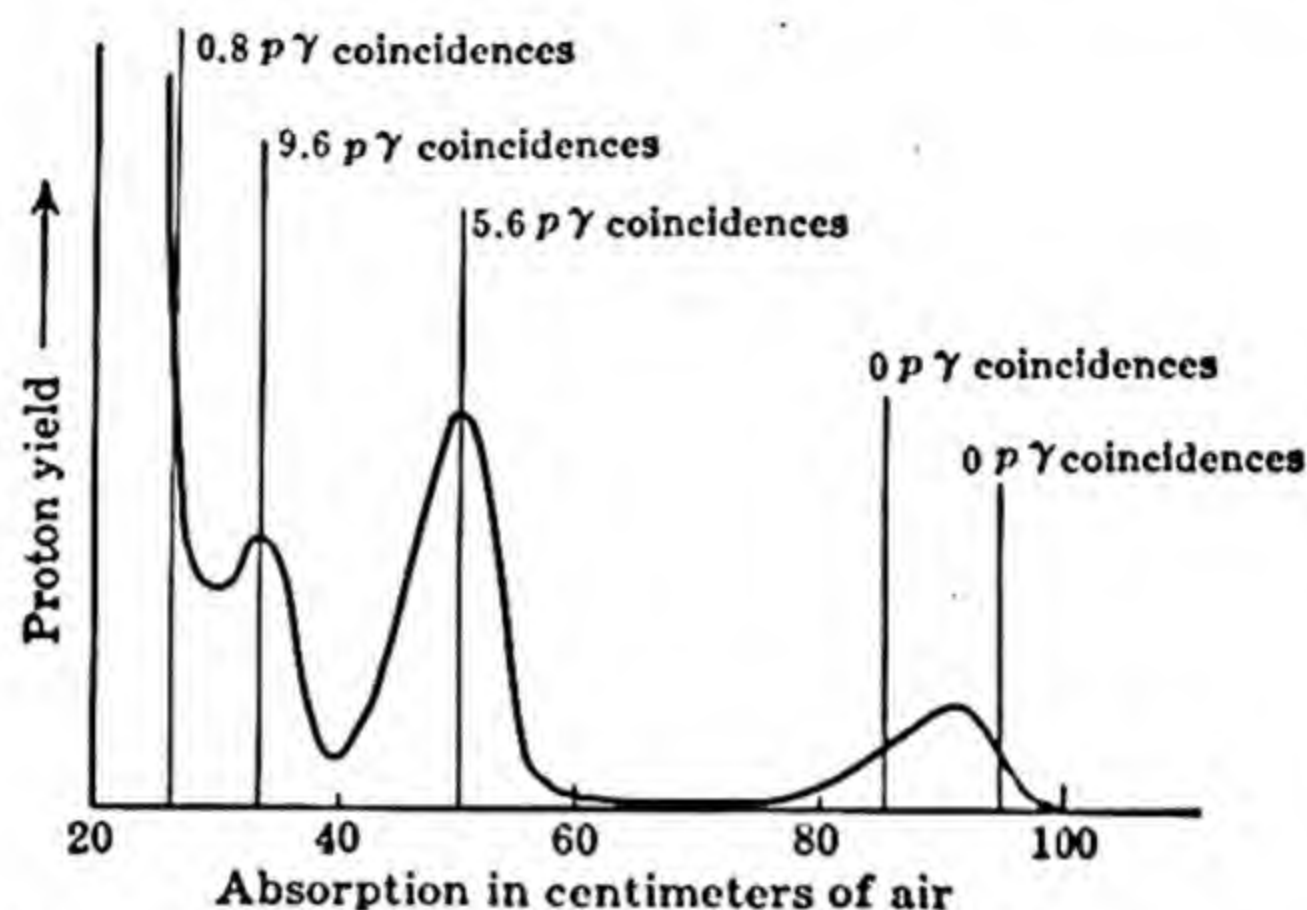


FIG. 10. Reproduction of an absorption curve taken by Benson for the  $\text{Al}^{27}(\alpha p)\text{Si}^{30}$  reaction. Four groups of protons can be seen. Those ending at about 100 cm leave the product nucleus of  $\text{Si}^{30}$  in the ground state, as is seen by the fact that no gamma rays are observed in coincidence. Those ending at 60 and 40 cm leave  $\text{Si}^{30}$  in the first and second excited states, and gamma-ray transitions remove the energy of excitation.  $p\gamma$  coincidences are observed, the number given being for 10,000 protons. The group ending at 25 cm is mostly due to recoil with hydrogen contamination, and only a few coincidences are observed.

heavy ionization of a slow particle (near the end of its path) by biasing the detection to count only such particles. An absorption curve obtained by Benson for the reaction  $\text{Al}^{27}(\alpha p)\text{Si}^{30}$  is shown in Fig. 10. The presence of three groups of protons is clearly seen. In order to prove that the process outlined in Fig. 8 is right a test can be made of the existence of gamma rays in time coincidence with the protons. The energetic protons should have no gamma rays associated with them, whereas the other groups should show coincidences. The coincidence rates per 10,000 protons are shown on the figure. It can be seen that the statement is verified.

A second absorption curve for the reaction  $\text{Al}^{27}(dp)\text{Al}^{28}$  taken by Wyly and Sailor and one of the authors is shown in Fig. 11. The existence of many levels is seen. These are plotted at the right of the



figure, where levels found by Seagondollar and Barschall in neutron cross section measurements for the same nucleus are included for comparison. It can be seen that energy levels are more closely spaced at high excitations.

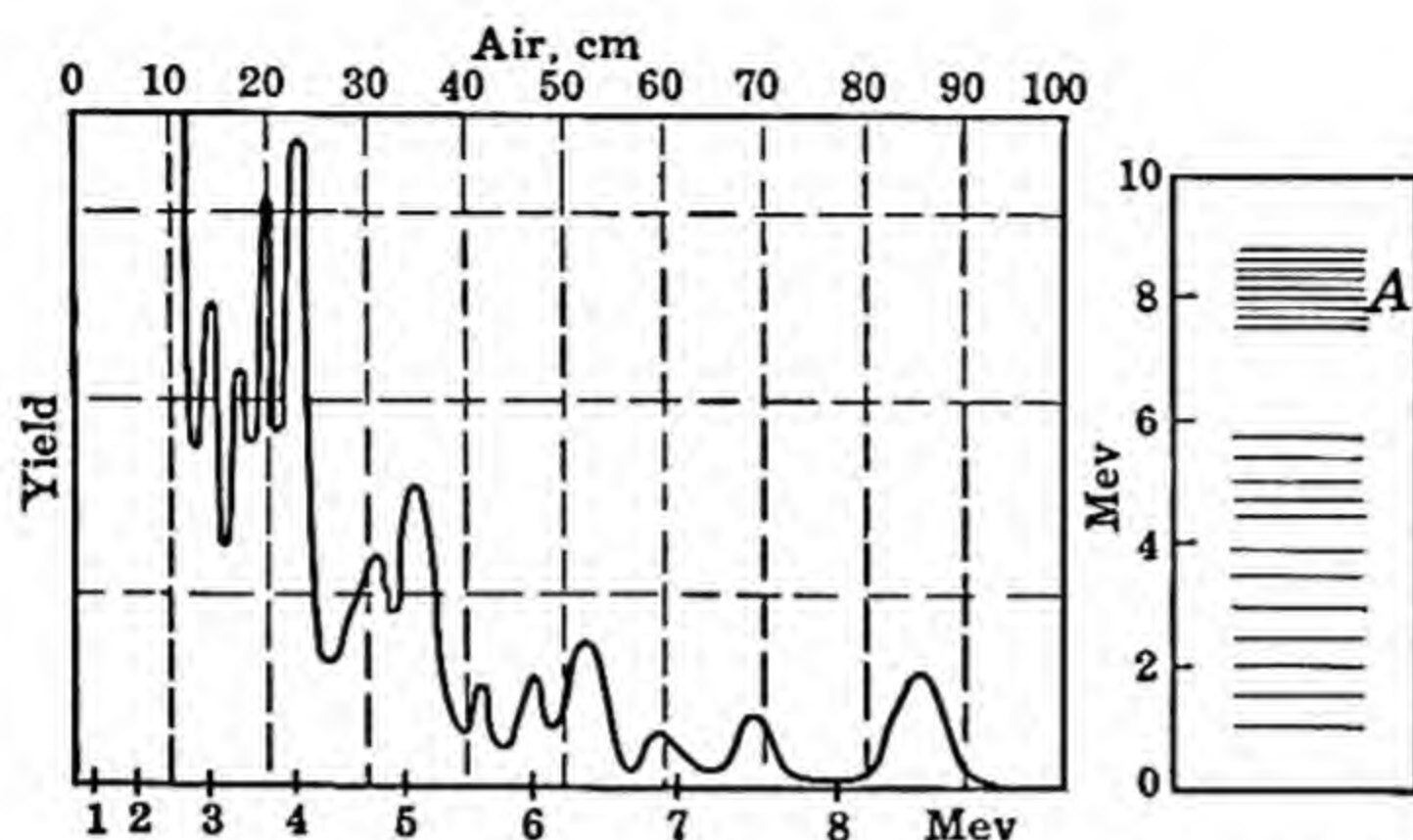
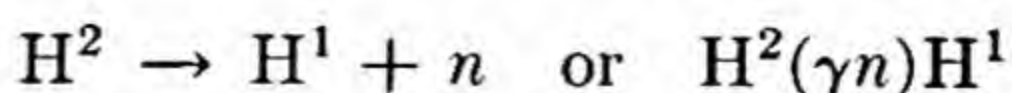


FIG. 11. Proton groups from  $\text{Al}^{27}(dp)\text{Al}^{28}$ , from data taken by Wyly and Sailor. The corresponding energy levels are shown on the right, with resonance levels observed by Seagondollar and Barschall at higher excitation shown at A. The density of levels is seen to increase with excitation.

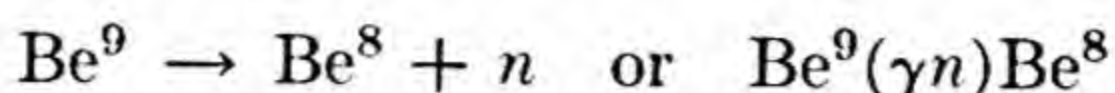
### Transmutation by radiation

The electromagnetic field of a gamma ray can act on the particles in a nucleus and cause a transmutation. This is the so-called *photodisintegration* process. While we were considering the *pn* type of reaction we called attention to the presence of a threshold energy which must be given to the proton before the reaction will go. This threshold feature is even more strongly present in photodisintegration, for the gamma ray possesses no mass of its own and must therefore achieve its results by sheer original energy. For this reason the process was discovered somewhat late. Ordinarily, if one takes any nucleus and removes any light nuclear particle from it, it will be found that the increase of mass needed to separate these two parts corresponds to about 9 Mev. This energy must be supplied entirely by the gamma ray, if it is to cause disruption. No such energetic gamma rays are found among naturally radioactive materials, and it was only by bombarding the relatively loosely bound deuteron that Chadwick and Goldhaber were able to show that this type of reaction could occur. They used the 2.6-Mev gamma ray from  $\text{ThC}'$  to bombard deuterium gas and found that protons were liberated. The process is



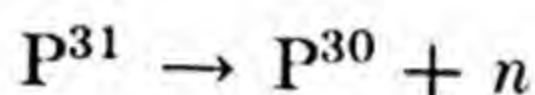


Beryllium can also be split up in this way, according to the reaction



These two reactions actually give rise to considerable numbers of neutrons, because the gamma ray is able to cause transmutations throughout a thick layer of material, as it is absorbed by a different process compared with charged particles. Such neutrons are of interest in that they are all of one energy, and they are therefore a useful tool in research. The energy is determined by the energy of the gamma ray which is used and the energy needed to separate a neutron from the target nucleus. It is unique, but it cannot be controlled.

The extensive series of photodisintegrations now known came as a result of the use, by Bothe and Gentner, of the energetic radiation from lithium bombarded by protons. In general there is so great a stray neutron background from this source of gamma rays that the neutrons cannot be detected directly. Frequently, however, the ejection of a neutron from a nucleus produces a radioactive element which can be detected and recognized. This was done by Bothe and Gentner in reactions such as



Here the  $\text{P}^{30}$  is radioactive, emitting a positron with a half-life of 3 minutes. The process of photodisintegration differs from other methods of transmutation in that it does not call on the interaction between particles to effect a change. The fundamental force that causes the disruption is the force resulting from the action of electromagnetic radiation on the particles in the nucleus. This force is not so great as the direct force between elementary particles, and the result is that the effectiveness of radiation is rather small. We do not, therefore, expect any great commercial use to be made of this reaction; it is rather to be useful in studying the nucleus itself.

A convenient source of neutrons can be made from radioactive antimony and beryllium. The energy of one of the gamma rays from Sb is 1.73 Mev, and this exceeds the threshold for photodisintegration of beryllium. The cross section for absorption of gamma rays in beryllium by the photodisintegration process is about 1 per cent of that for absorption by electrons, so that about one gamma ray in a hundred produces a neutron. This gives a useful neutron source of low-energy neutrons.



High-energy radiation which is very suitable for producing photodisintegrations can be produced by the impact of the electron beam of a betatron or synchrotron on a target. The radiation so produced is continuous in energy but has a definite maximum which is fixed by the electron energy. The precise study of photodisintegration by this means is thus rather hard, but it has proved possible to measure the threshold energy at which certain reactions can take place. That the reaction has occurred is made evident by the formation of the proper kind of radioactivity. The threshold is found by varying the beam energy until this just takes place. Such thresholds are very useful in assigning mass values.

### High-energy bombardment

The fact that projectile energies above 100 Mev are becoming commonplace is causing a new field of study of high-energy reactions. These differ considerably from the simple processes described so far in that the availability of enough energy is guaranteed and the question is one of the distribution of the products of the reaction. For example, if any target is bombarded by 200-Mev deuterons and then examined for radioactivity there at once appears a rich variety of half-lives, and the use of chemical separations shows that the breaking off of several particles at once (a process called spallation) takes place readily. The general rule that any nuclear reaction which is energetically possible will take place seems to be obeyed, though the probabilities of various processes are different. The careful study of the breaking up of nuclei under high-energy bombardment will probably contribute greatly to the understanding of the nucleus. However, it is doubtful whether the manufacture of radioactive elements for tracer purposes will commonly be carried out by this method.

### Transmutation by mesons

The negative  $\pi$ -meson seems to have displaced the neutron in order of potency for reaction. This is because the meson is not even indifferent to the coulomb field: it is attracted by it. An energetic meson, moreover, is not heavily ionizing and therefore does not squander all its energy in moving electrons in the outer atom. These negative mesons are readily absorbed by nuclei and as they bring with them 142 Mev of mass energy the resulting process is an explosion. Since most of the observations of this kind of process have been made in cloud chambers or photographic plates the appearance of such an explosion looks like a star and the process is now called "star produc-



tion." A picture of a negative meson producing a star is shown in Fig. 12.

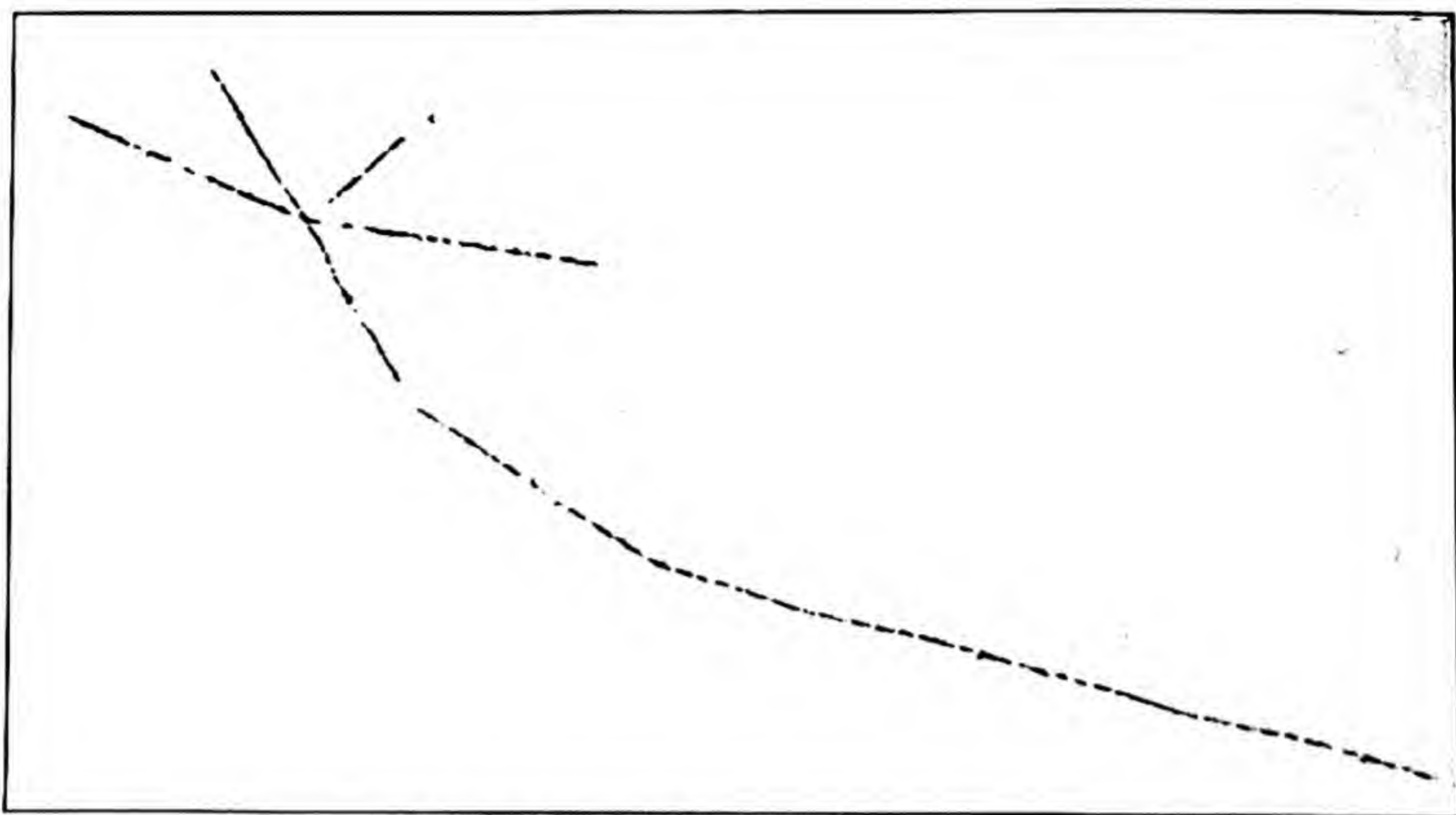


FIG. 12. Star produced by a negative  $\pi$ -meson. The track is recorded in photographic emulsion. The meson is scattered twice as it traverses the emulsion and finally enters a nucleus producing a star. (Picture due to Dr. E. O. Salant.)

## Conclusion

This chapter bids fair to be the longest in the book. It is an interesting chapter to write, but rather unsatisfactory when read over. The truth is that the important ideas in nuclear chemistry, like the importance of mass and energy, can be said rather quickly, but that the "feel" of the subject cannot be imparted so soon. It takes thought and much discussion to become happy at the array of possibilities now presented to us. It is intended that this chapter serve, not as a compendium to all nuclear reactions, but as an ice breaker to enable the reader to join in discussion, or read the more thorough summaries either now available or shortly to be written.

There is much valid witticism at the expense of summarizing summaries. Nevertheless, we are going to summarize this chapter. As a beginning we show the table, in which the various types of reaction are listed together with the salient points of each.

The progress of a reaction is first of all conditional on a favorable mass balance; the mass of the two reacting particles plus the mass equivalent of the kinetic energy of the projectile must exceed the mass



SUMMARY OF REACTION TYPES

Reaction Type	Normal Mass Change	Dependence on Energy of Projectile	Yield	Type of Radioactivity Usually Produced	Sample Reactions
$n$ capture	Positive	Resonance	Virtually 100%	Electron	$\text{Ag}^{107} + n \rightarrow \text{Ag}^{108}$ $\text{Br}^{79} + n \rightarrow \text{Br}^{80}$
$np$	Slightly positive	Smooth	Large for light elements; escaping barrier to consider	Electron	$\text{N}^{14} + n \rightarrow \text{C}^{14} + \text{H}^1$ $\text{S}^{32} + n \rightarrow \text{P}^{32} + \text{H}^1$
$n\alpha$	Slightly positive in light elements; negative in heavy	Smooth	As above	Electron	$\text{F}^{19} + n \rightarrow \text{N}^{16} + \text{He}^4$ $\text{Al}^{27} + n \rightarrow \text{Na}^{24} + \text{He}^4$
$n, 2n$	Very negative	Smooth	Small	Positron	$\text{N}^{14} + n \rightarrow \text{N}^{13} + 2n$ $\text{P}^{31} + n \rightarrow \text{P}^{30} + 2n$
$p$ capture	Positive	Resonance	Large	Positron	$\text{C}^{12} + \text{H}^1 \rightarrow \text{N}^{13}$ $\text{F}^{19} + \text{H}^1 \rightarrow \text{Ne}^{20}$
$pn$	Negative	Threshold: smooth, increasing with energy	Large	Positron	$\text{B}^{11} + \text{H}^1 \rightarrow \text{C}^{11} + n$ $\text{O}^{18} + \text{H}^1 \rightarrow \text{F}^{18} + n$
$p\alpha$	Slightly positive in light elements; negative in heavy	Smooth, increasing with proton energy	Large	Generally stable products	$\text{F}^{19} + \text{H}^1 \rightarrow \text{O}^{16} + \text{He}^4$ $\text{Al}^{27} + \text{H}^1 \rightarrow \text{Mg}^{24} + \text{He}^4$
$pd$	Very negative	Smooth as above	Small	Only one case established	$\text{Be}^9 + \text{H}^1 \rightarrow \text{Be}^8 + \text{H}^2$
$an$	Slightly negative in light elements; positive in heavy	Smooth	Large for elements where barrier penetration is easy	Positron	$\text{B}^{10} + \text{He}^4 \rightarrow \text{N}^{13} + n$ $\text{Al}^{27} + \text{He}^4 \rightarrow \text{P}^{30} + n$
$ap$	Slightly positive except some light elements	Smooth	As above	Generally stable products	$\text{Al}^{27} + \text{He}^4 \rightarrow \text{Si}^{30} + \text{H}^1$ $\text{N}^{14} + \text{He}^4 \rightarrow \text{O}^{17} + \text{H}^1$
$dp$	Always positive	Smooth	As above	Electron	$\text{Na}^{23} + \text{H}^2 \rightarrow \text{Na}^{24} + \text{H}^1$ $\text{P}^{31} + \text{H}^2 \rightarrow \text{P}^{32} + \text{H}^1$
$dn$	As above	Smooth	As above	Positron	$\text{C}^{12} + \text{H}^2 \rightarrow \text{N}^{13} + \text{H}^1$ $\text{Be}^9 + \text{H}^2 \rightarrow \text{B}^{10} + \text{H}^1$
$d\alpha$	Always positive	Smooth	As above	Generally stable products	$\text{O}^{16} + \text{H}^2 \rightarrow \text{N}^{14} + \text{He}^4$ $\text{Al}^{27} + \text{H}^2 \rightarrow \text{Mg}^{25} + \text{He}^4$
$\gamma n$	Always negative	Sharp threshold	Small	Positron	$\text{Be}^9 + \gamma \rightarrow \text{Be}^8 + n$ $\text{Br}^{81} + \gamma \rightarrow \text{Br}^{80} + n$
$\gamma p$	As above	As above	As above	Electron	$\text{H}^2 + \gamma \rightarrow n + \text{H}^1$
$-\pi$ capture	Positive	Smooth	100%	Star formation	

CONCLUSION



of the resulting products. Second, all charged particles must either surmount or penetrate through a potential barrier which becomes higher and thicker the greater the atomic number of the target element. Third, there will always be a competition among various possible reactions so that one cannot always predict the nature of the yield given. On these counts neutrons and deuterons are on the whole the most favorable projectiles for causing transmutations, although this statement must not be interpreted as meaning that they are always so. Roughly speaking, if there is no unusual competition, the yield of changed nuclei from bombardment by any particle which has enough energy to overcome the barrier will be one for every 100,000 incident particles. This figure can vary by as much as a factor of 10, and it applies only to a thick target. As the energy of the bombarding particles becomes higher with technical advance, it is likely that many new varieties of reaction will be discovered, rendering this long chapter a slender first section of a large subject. It is prophesied, however, that these reactions will be of little use in manufacturing radioactive isotopes for practical work.

We hope that a reasonable digesting of this chapter will enable the reader to master the subject of radioactivity and the production of radioactive materials, the subject of Chapter 6.

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## 6. Radioactivity

*. . . such interchange of state,  
Or state itself confounded to decay.*

Radioactivity is a veritable godsend to the physicist. He is used to expatiating on a subject whose main attraction is the one thing most people resent—exactness—which seems to the listener like being pinned down all the time, and radioactivity, though it has been imbued by the press with all the mystery of relativity, is actually mostly a simple descriptive subject. The basic ideas are easy, and it takes very little familiarity with the principles to be able to predict the general behavior of any given radioactive element; only when the actual process is closely examined does the theory become unpleasant. For practical purposes such theory is not necessary.

### Radioactivity in general

In the first chapter we described the extreme simplicity of the scheme of nature. Neutrons, protons, electrons, and forces—no more. We also asked and for the moment answered the question as to the way in which we could mix neutrons and protons in a nucleus. We can mix them how we like, but nature sees to it that, after the mixture is made, the neutrons or protons interchange until the mixture is one of her own recipes. These recipes have been standard for more than a billion years. The process of readjustment is known as “radioactivity.”

Let us now take a closer look at the events taking place; in fact, let us concentrate on one example for a while. The simplest radioactive element is extra heavy hydrogen or tritium,  $H^3$ . This element was thought for a long while to be stable, but a brilliant piece of investigation by Alvarez showed that, in spite of the form book,  $H^3$  is the wrong horse to back.  $He^3$  is the winner. In more detail, the combination of two neutrons and one proton is less stable than the combination of two protons and one neutron. This, nevertheless, does not



prevent us from being able to make a considerable number of atoms of  $H^3$ ; for example, the reaction



is one of the most prolific known, and we can easily build up many millions of  $H^3$  atoms.

Consider for a moment one of them. It is unstable. It wants to become  $He^3$ , which it can do at any time by changing a neutron into a proton and emitting an electron. This process is not quite so simple as it sounds, for the neutron has a certain tendency to remain a neutron, and if the difference in stability between  $H^3$  and  $He^3$  is not very great the  $H^3$  will remain such for a considerable time; but in the end it will go over. *This process, the passage from one nearly stable nucleus to one which is stable, is the underlying process of radioactivity. Every radioactive element is one which is off the beaten track; it is a freak, but a freak with the power to correct itself by changing its nature so that it returns to type.*

We can illustrate this account of radioactivity further by considering various types of carbon. In order to have a carbon nucleus only one real requirement is involved: the nuclear charge must be 6, or six protons must be present. The commonest form of carbon has six neutrons as well, making a total of twelve particles. Now let us imagine that a neutron is added; we still have carbon, but is it stable? The answer is yes, although it is not so abundant as the ordinary form. Very well, add a second neutron. We now have  $C^{14}$ , and we quickly perceive that a stable element having the same number of particles (fourteen), namely  $N^{14}$ , already exists. We therefore confidently predict that  $C^{14}$  is a freak, that it will adjust its constitution until it becomes normal, and that therefore it will change one of its neutrons into a proton, emitting at the same time an electron. In other words, we predict that  $C^{14}$  is beta-radioactive. Such is the fact.

Instead of adding neutrons we could have imagined them removed. Then on removing one neutron from  $C^{12}$  we obtain  $C^{11}$ , and again we see that the number, eleven, of particles is already taken up by stable  $B^{11}$ . We therefore predict that  $C^{11}$  is radioactive, that it will convert a proton into a neutron with the emission of a *positron*. And it is so. In the same way it can be seen that  $Na^{24}$  usurps the number occupied by  $Mg^{24}$  and so  $Na^{24}$  is radioactive; that  $P^{32}$  will decay into  $S^{32}$ , and so on. In fact, it becomes clear that, once the technique of bombardment described in the last two chapters has been developed, the num-



ber of radioactive nuclei produced will be very great, actually greater than the number of stable nuclei, but that the general nature of radioactive processes will be as simple as we have already described.

If the reader will stop here for just a moment to consider what we have said regarding the nature of radioactivity, he will see that the major idea is very simple: as simple as rolling off a log, which is after all a passage from a nearly stable state to one more stable. *This major idea need not bother him*, and it is in reality the secondary questions which now arise that may cause him trouble. Such questions concern the actual time taken to effect the change and the energy of the products of the change. We can now consider radioactivity in more detail.

### Statistical nature of atomic theory

Before we look more closely at the process of reversion to type it will repay the effort to consider for a moment the rules which have been found to apply to small-scale matter, such as atoms or parts thereof, and then see how they will apply to radioactivity. In the first place it has been found that it is impossible to describe the motion of electrons or protons or any small enough particles in detail. Notice, by the way, that it is not necessary to do so, for we are nearly always compelled to deal with vast aggregates of atoms, and the motion of one alone need not be followed. If we cannot follow the behavior of an electron in detail, the question arises as to what we can do; and the answer is that we can calculate a quantity which will evaluate the chance that an electron be at a given place at a given time, although we cannot calculate its precise path. The quantity we calculate is quite accurate and of the greatest value; in fact, it is all we need; but it must be remembered that it informs us of only a probability, not a certainty.

A little digression will show the difference between the modern outlook and what our large-scale knowledge led us to expect until this century. A careful study of Harvard men has shown that their average height is greater than that of the rest of the population. This means that if a man is known to be a Harvard man he will be expected to be taller than average, but there is no reason why any one Harvard man will be tall. In fact, if we were going to use this as a test for whether a man were a Harvard man, we would have to admit that it would not help much. On the other hand if we were presented with a party of a thousand Harvard men and asked to verify whether



they were genuine, we could get real information by finding their average height and seeing if it were above the average for the country. The difference between the two outlooks can be seen from our illustration. Modern atomic physics is of service only in the statistical sense; it can give real information only when a large number of cases is considered, whereas the course of physical discovery up to the present century led us to expect that in each case we could follow what happened accurately if we only had the right theory to help us to explain the events.

Let us repeat, then, that to describe the motion of an electron or any part of an atom we must calculate the value of a certain function at each place and then its value will tell us what the chance is that an electron will be at that place. One interesting and exciting result that follows from this kind of procedure is that in an atom an electron can have only a limited number of values of energy, the "energy levels" of the atom, and these energy levels play a vital role in spectroscopy, which has justified the method of procedure of modern physics so convincingly.

### Probability and radioactivity

To return once again to radioactivity. A neutron and a proton are supposed to be two states of the same thing, with the proviso that a neutron may change into a proton and liberate an electron and neutrino at the same time. Now we may suppose that the change takes place because some force acts tending to make it occur. We do not yet presume to suppose the nature of the force; nuclear physics is still a young science; but we suppose that there is such a force and that it tends to make such a change. In a stable nucleus there is a balancing of these forces so that there is no net force seeking to make the change, whereas in an unstable nucleus there is a net unbalanced force which is tending to convert a neutron into a proton. All we have to do is to calculate the value of the function corresponding to the changed neutron, and this will tell us what the chance is that, at a given time, a neutron will have become a proton and a radioactive change will have taken place. Notice that all we get is the *chance*. We get no definite information about when a single atom will change. *All we can say is that there is a certain chance within a given interval of time that the neutron will take the alternative form of a proton, electron, and neutrino, and, of course, the greater this chance the more rapid is the process of radioactive decay.*



## Radioactive decay

Before we go on to consider the predictions of the theory of nuclear instability it is important to look at the main features of the manner of change just described above—a manner in which there is a certain constant probability of a change taking place in a given interval of time. Understanding of the nature of this apparently simple way of change will go a long way to help in understanding the whole nature of radioactivity.

Let us therefore first consider the meaning of the word “probability.” If a certain number  $N$  of cases be considered, the probability of a certain distribution of cases is the ratio of the number in that category (say  $n$ ) to the total number  $N$ , that is  $n/N$ . Thus, if we toss pennies  $N$  times, the probability of getting heads is the number of times we find heads divided by the total number of times  $N$ .

Now consider the changing of a million atoms. Suppose that during 1 second there is a probability of radioactive change of  $\frac{1}{1000}$ . This means that in any 1 second a fraction  $\frac{1}{1000}$  of the atoms will have changed. Then 1 second after the beginning of our study of the million atoms we expect that we shall find 1000 atoms in a changed condition. This change, once made, is complete, and we are now in possession of only 999,000 atoms. (The change from stable to unstable cannot occur as there is not the energy available to make it go.) In the next second the probability is still the same, but as we now have only 999,000 atoms a slightly smaller number will change: 999, to be precise. This leaves us with 998,001 atoms, and therefore in the next second 998 atoms will go over. (Not exactly, as there is the odd one to consider, but this means only that it is likely that 998 will go.) The reader does not need to continue with this form of arithmetic; he can see that, if we let  $N$  be the number of “parent” atoms left and  $n$  the number changing in a second, then we have the relation

$$n = \frac{1}{1000} N$$

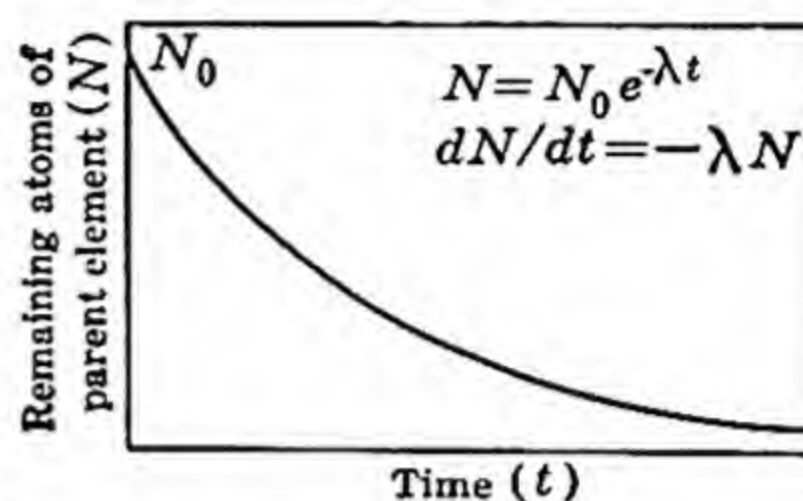


FIG. 1. Graph representing the number of atoms of a radioactive element left after different intervals of time. This is equivalent to a “decay curve.” The curve corresponds to the relation  $N = N_0 e^{-\lambda t}$ , which is the same as  $dN/dt = -\lambda N$ , meaning that the rate of change is proportional to the number of atoms of the decaying element.



If we plot a graph of the number of atoms of the unstable or "parent" atoms left after different intervals of time it will appear somewhat as indicated in Fig. 1. The number diminishes rapidly at first, and then more slowly, approaching but never reaching zero after a long interval.

This description in terms of probability can be made numerical. Let  $-dN$  be the number of atoms changing due to the radioactive process in a time  $dt$ . The negative sign is used because the number is diminishing. Then we have for the probability  $-dN/N$ , and if this is to be proportional to  $dt$  we have

$$-\frac{dN}{N} = \lambda dt$$

or

$$\frac{dN}{dt} = -\lambda N \quad (1)$$

The relation giving  $N$  explicitly is

$$N = N_0 e^{-\lambda t} \quad (2)$$

where  $N_0$  is the initial number of atoms at the arbitrary starting time. The quantity  $\lambda$  is called the *disintegration constant* or *decay constant*. For ordinary use we can rewrite equation 2 by taking the logarithm of both sides, with the result

$$\ln N_0 - \ln N = \lambda t \quad (3)$$

This is one of the most important equations in applied nuclear physics. It should be remembered that natural logarithms are used above, and that if common logarithms are used experimentally the value of the decay constant deduced without correction will be too small by the factor 2.30.

Equation 3 refers to the number of atoms of the parent element present, which is not usually measurable. It is far easier to observe the effects due to the radiations emitted during the actual change; this would mean observing an effect proportional to the *rate of decay*,  $dN/dt$ . It follows from equation 1 that observation of  $dN/dt$  also gives us  $-\lambda N$ , so that we are observing a quantity which is proportional to the number of atoms present. If we put  $N = (-dN/dt)/\lambda$  and  $N_0 = (-dN_0/dt)/\lambda$  in equation 3 we get

$$\ln \frac{dN_0}{dt} - \ln \frac{dN}{dt} = \lambda t \quad .$$



or if we put  $A_0$  and  $A$  for the "activities" at the initial time and any time, respectively, these activities are directly proportional to  $dN_0/dt$  and  $dN/dt$ , so that we get the final relation

$$\left. \begin{aligned} \ln A_0 - \ln A &= \lambda t \\ \text{or in common logarithms} \\ \log A_0 - \log A &= \frac{\lambda}{2.30} t \end{aligned} \right\} \quad (4)$$

In Fig. 2 is shown the type of line resulting from plotting the common logarithm of the ionization current produced in a Lauritsen electro-scope by a source of radiosodium against the time in hours. The sloping straight line can be seen to verify relation 4.

Although the most direct way of describing the decay of a radioactive element would be to give the value of the decay constant  $\lambda$ , this is not commonly done. It is more usual to describe it by giving the time required for the activity to fall to half its initial value. This half-life is the quantity usually given in tables of reference. Its relation to the decay constant is easily seen from equation 4. If we put the value of  $A$  as  $A_0/2$  when the time is  $t_H$ , we get  $\ln 2 = \lambda t_H$ , or  $t_H = \ln 2/\lambda$  or  $0.693/\lambda$ . A convenient equation involving half-life is  $A_0/A = 2^{t/t_H}$ , which enables quick calculation.

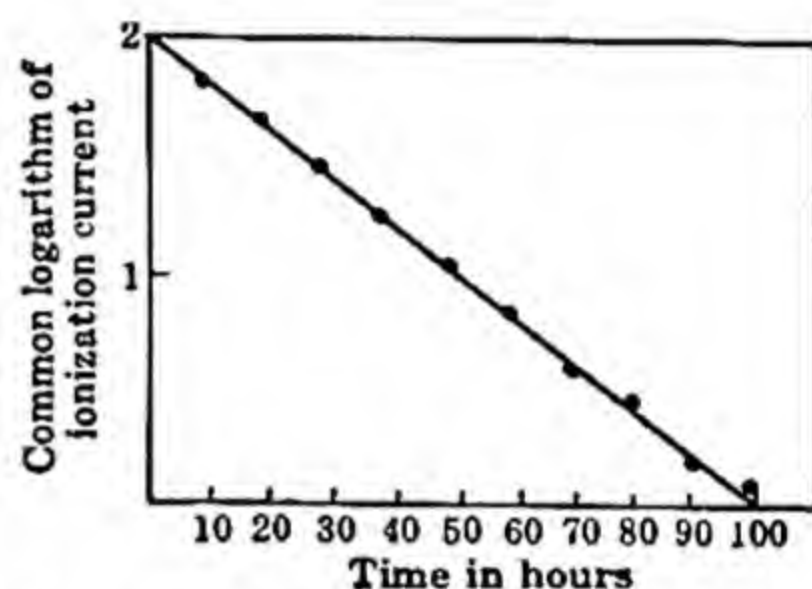


FIG. 2. Graph of the common logarithm of "activity," in this case ionization current produced in a Lauritsen electro-scope by a radiosodium source plotted against the time in hours.

To obtain the half-life from a logarithmic plot as in Fig. 2, we do not need to calculate the decay constant, but merely find the time elapsed for the logarithm of the activity to fall by  $\log_{10} 2$ , or 0.301. Perhaps there is no harm in pointing out that it is quite immaterial where we choose the initial time; no matter how many atoms have decayed, the decay constant is the same for the remainder.

### Energy of the products of radioactive decay

The second feature of radioactivity which needs treating in detail is the energy of the products emitted. We have so far indicated that the process of radioactivity is the passage from an unstable to a stable nucleus, and we have said that this takes place by the conversion of a neutron into a proton. It can easily happen that the unstable nucleus



attains stability by the conversion of a proton into a neutron; this often happens, and there is no essentially new feature about this method of adaptation except that the proton becomes a neutron and a positron. The emitted particle is then a positron and differs from an electron only in charge and in the fact that its production requires the using up of 1 Mev merely to create the positron. This need for an initial 1-Mev "starter" has interesting consequences which will be discussed later in the chapter. Whichever method of decay is found to take place it is still of interest to consider the energy of the emitted electron or positron.

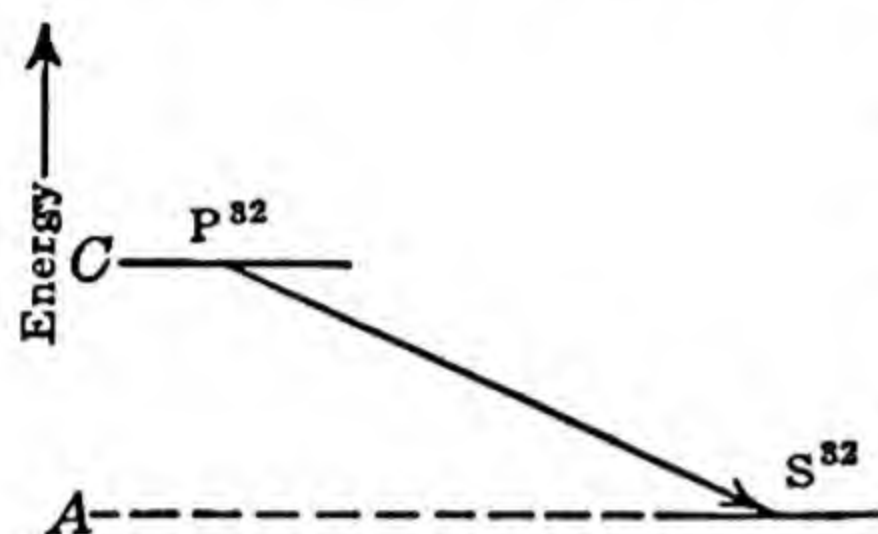


FIG. 3. Energy levels for  $P^{32}$  and  $S^{32}$ . The energy difference represented by  $AC$  is the energy available as kinetic energy for the electron, the neutrino, and the recoil atom.

A good way to see the process from the viewpoint of energy is to consider two energy-level diagrams for the initial nucleus and the final stable product. In Fig. 3 we have represented the nucleus of  $P^{32}$  and the stable nucleus  $S^{32}$  next to it. The upper level represents the unstable  $P^{32}$ , and the lower, the stable  $S^{32}$ . When the radioactive change takes place there is a release of energy represented on the diagram by the height  $AC$ . This energy when released causes motion; the distribution of the speeds of the particles concerned

(which we at present suppose to be the daughter atom  $S^{32}$ , and the emitted electron) will be determined by the conservation of momentum. Since the mass of the atom of  $S^{32}$  is so much greater than that of the electron we expect the speed of the electron to be greatly in excess of that of the "recoil atom," and this means that virtually all the energy set free is given to the electron. The picture we have drawn leads us to predict that the radioactive decay of  $P^{32}$  will take place with the emission of electrons of one energy, that energy being a little less than the energy available from the difference in mass between  $P^{32}$  and  $S^{32}$ . Actually this is not so. *It is found that electrons having all energies up to the maximum value are emitted.* The maximum value is that which we would have expected to appertain to all the electrons, but it alone gives the necessary balance of energy.

The explanation of this rather disconcerting fact has been given by Fermi, following a suggestion by Pauli. Fermi proposed that in addition to the electron a new particle, the "neutrino," having properties rather like a very light neutron, is emitted. This neutrino is hard to



detect as it does not produce any effect on our measuring apparatus, but at the same time it is capable of carrying off energy. According to the Fermi theory the total disintegration energy, the energy equivalent to the mass difference between parent and daughter elements, is given as kinetic energy to the recoil atom, the electron, and the neutrino. Since the mass of the last two is so small relative to the mass of the first, they absorb nearly all the energy. Then if the electron takes all the energy and the neutrino none, the measured energy of the electron will be the disintegration energy. This fits the experimental observations. On the other hand it is also possible that the neutrino takes all the energy, in which event the electron would not even be detectable. The emission of very low-energy electrons is actually found. It is clear that the chance of either of these extremes is rather small and that somewhere in between, where there is rough equality of energy, the chance will be much greater.

It is of practical importance to have some idea of the relative numbers of electrons with energies at various values short of the maximum. This information is given accurately by the Fermi theory, but as this involves relativistic quantum theory it is hard to give a simple explanation here. The result, when obtained, is rather complex, and it is not easy to compile a set of simple tables which will enable the distribution to be calculated in a given case.

The neutrino and electron between them take almost all the energy; only a little is taken by the recoiling nucleus. Sherwin has recently shown that the measurement of this energy fits with the neutrino hypothesis. The rest mass of the neutrino is very small or zero, and this results in a slight excess of the total energy going to the neutrino. The most probable energy for the electron is accordingly about one-third the maximum.

The distribution curve for the electrons from  $P^{32}$  is shown in Fig. 4. A distribution curve is one in which the number of electrons having

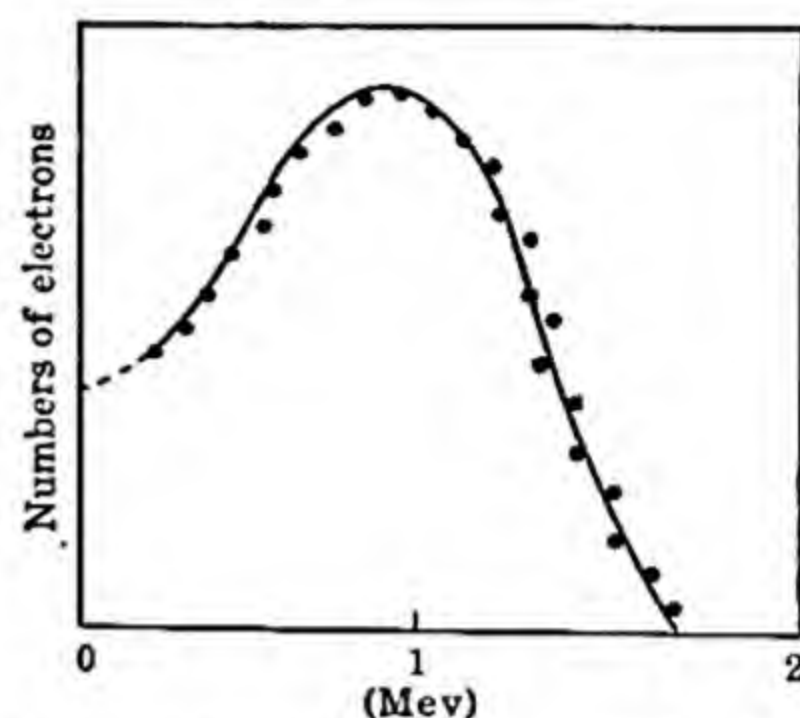


FIG. 4. Experimental distribution curve obtained by Lawson for the electrons from  $P^{32}$ . A roughly symmetrical curve is found, which agrees with the Fermi theory, indicating that the electron and neutrino, on the average, share the energy equally. The upper limit of 1.69 Mev corresponds to the difference in mass between  $P^{32}$  and stable  $S^{32}$ .



energies between two limits is plotted against the average energy of the limits. If such a curve is plotted by observing the curvature of the electron tracks in a cloud chamber placed in a magnetic field, the observer will measure the curvature of, say, a thousand tracks, express the curvatures in terms of energy, and then lay out a dozen groups or so, with energies perhaps 200,000 electron volts apart. He then counts the number of tracks in each interval and plots the values against the appropriate average energy, so producing a distribution curve. The best curves are usually obtained with a so-called beta-ray spectrometer, in which the beta rays are bent through a slit system by a magnetic field and then detected by a counter.\* In the pioneer stages of the study of these distribution curves the cloud chamber placed in a magnetic field has the advantage that all the tracks can be seen and individually considered to be sure that they have the assigned energy; after the ground has been broken the spectrometer is more accurate as it allows the measurement of a much larger number of particle energies. We have mentioned this fact as it is so for almost all applications of the cloud chamber. It yields sure data but not a very large sampling. To return to the distribution curve itself. For experimental reasons the curve is incomplete near the region of zero energy, but the remainder shows a reasonably symmetrical distribution about

\* We do not intend to explain the precise method of determining distribution curves, but, as the reader may consult original work for his own purposes, we feel that a word of explanation of the rather confusing terminology is in place. It is usual to describe the energy of the electron in terms of its curvature in a magnetic field. If we consider a slow electron of velocity  $v$ , then this is equivalent to a current element  $idl$  of value  $ev$ , and in a magnetic field there will be exerted a force  $Hev$  perpendicular to the direction of motion of the electron, where  $H$  is the magnetic field intensity. This produces an acceleration perpendicular to the direction of motion and so causes the electron to follow a circular path of radius  $\rho$ , such that the applied force equals the mass times the acceleration due to the circular motion. In symbols this is:

$$Hev = \frac{mv^2}{\rho} \quad \text{or} \quad v = \left(\frac{e}{m}\right)(H\rho)$$

Thus the quantity  $H\rho$  is proportional to the velocity of the electron. At high speeds the mass of the electron is greater, but the quantity  $H\rho$  still measures the momentum of the electron. It is therefore not an inconvenient way of describing the momentum, especially for experimenters in this field. A conversion formula to Mev is

$$H\rho = \frac{10^4}{3} [E(E + 1.02)]^{1/2}$$

where  $H$  is in gauss,  $\rho$  in centimeters, and  $E$  in Mev.



a mean value of 0.85 Mev, and the maximum energy has the value 1.69 Mev, almost exactly double. It is of importance to notice that there is considerable loss if for any reason a counter or electroscope detects only high-energy electrons, for the numbers fall off rapidly once one has passed 80 per cent of the maximum. This matter of the detection of electrons will be treated fully later.

If we now agree that the bell-shaped distribution curve is the expected type for a simple example where there is only one possible energy release, we have to inquire into the nature of the distribution which holds where the unstable nucleus can change into one or more excited states of the product nucleus. In Fig. 5 we show the levels for  $\text{Cl}^{38}$  and stable  $\text{A}^{38}$ . It happens that  $\text{A}^{38}$  has two excited states to which  $\text{Cl}^{38}$  could go. Referring to the figure we see that in addition to the transition marked (1), or ground state to ground state, there are two other possible transitions marked (2) and (3), or ground state to first and second excited states. It is found that each of these transitions has its own bell-shaped distribution curve, and the complete curve is the composite of the three. The total numbers in the separate bell-shaped curves are not easily predictable, but it often happens that the numbers occurring in the curves of less energy are greater. It is not impossible that in actual fact there are many cases of composite curves which have not yet been adequately analyzed, so that this rule must be taken as not finally established.

In Fig. 6 we show the experimentally obtained distribution curve for  $\text{Cl}^{38}$ . It can be seen that the general shape is different from that of  $\text{P}^{32}$  and that we can draw in the three curves as indicated and regard the actual distribution as due to the superposition of the three. The three curves have maximum energies at 4.9, 2.8, and 1.2 Mev, respectively, and these figures mean that the energy available in each transition is one of three discrete values; these values correspond to the transitions given in Fig. 5. We can therefore deduce that the

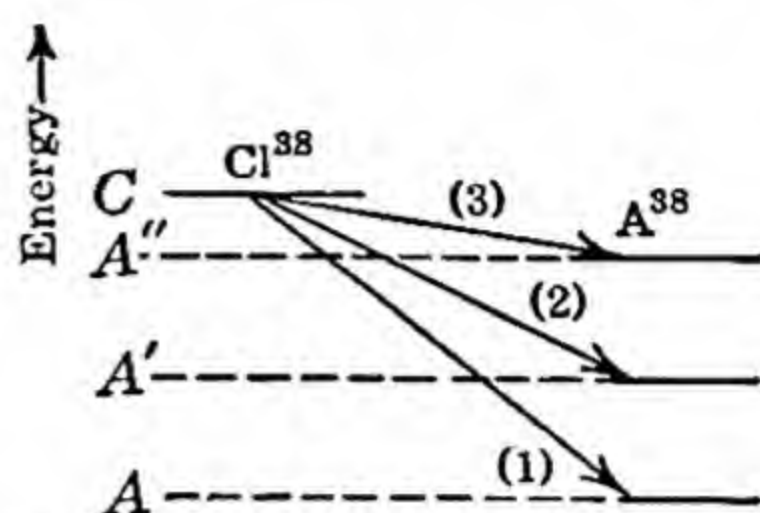


FIG. 5. Energy levels of unstable  $\text{Cl}^{38}$  and stable  $\text{A}^{38}$ .  $\text{Cl}^{38}$  decays into  $\text{A}^{38}$  by emitting an electron and a neutrino. If the transition is to the ground state as in (1), then the electron and the neutrino have the full energy corresponding to the difference  $AC$ . If the transition is to the first excited state, the electron and the neutrino take the energy  $A'C$  and the excitation energy  $A'A$  is emitted as gamma radiation. A transition to the second excited state will mean a third group of electrons and the subsequent emission of the excitation energy  $A''A$  as one or two quanta.



first excited state of  $A^{38}$  is  $4.9 - 2.8$  Mev above ground level. That is 2.1 Mev. The next state of excitation is  $4.9 - 1.2$  Mev, or 3.7 Mev above ground.

Now, if this interpretation is right, the fact that the residual  $A^{38}$  is often left in an excited state must mean that at some later stage it will emit a gamma ray and return to ground level. This actually occurs, and as it is impossible for a nucleus to remain excited for longer than about  $10^{-13}$  second (unless the degree of excitation is very small,

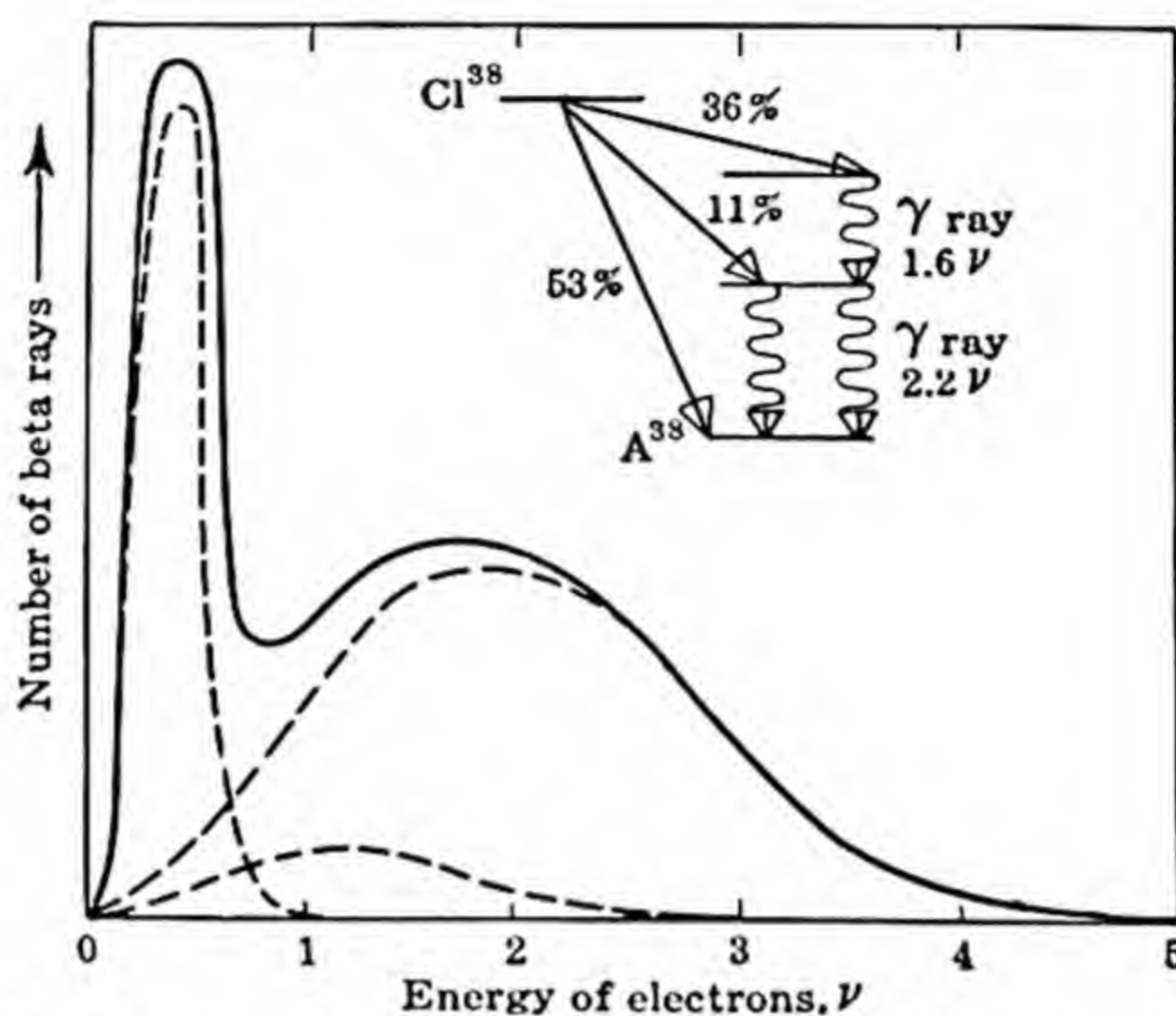


FIG. 6. Energy distribution of the electrons from  $Cl^{38}$  which decays to  $A^{38}$  in both the ground state and two excited states. The overall curve, which is derived from measurements made by K. Siegbahn, can be analyzed into three distributions having maximum energies at 4.9, 2.8, and 1.2 Mev. The percentage taking part in each transition is given in the insert.

or some great change of spin is involved), the gamma ray is emitted virtually simultaneously with the beta ray. This fact has led to some interesting work in which this coincidence in time between the gamma ray and the electron has been tested by a coincidence method of counting as described in Chapter 3. The coincidence is verified, but the unexpected result is found that frequently there is also coincidence between an electron which apparently has the energy of a transition to the ground state, and a gamma ray. This means that the numbers in the true group of electrons which correspond to the ground-state transition are so small as to escape detection and lends support to the suggestion that the lower-energy groups are favored. In concluding this paragraph it should be pointed out that where the beta rays corresponding to such groups as these are considered, where one level of



the parent atom decays to various levels of the product, all the beta rays decay with the same period. This is because a definite fraction of the electrons leaving the parent nucleus go to each of the levels of the final nucleus so that the decay constant for all is the sum of three decay constants and adds up to one single value. If the subsequent adjustment in the final nucleus takes longer than the beta-ray transition then there may appear a second half-life. This is not common.

### Relation between disintegration energy and half-life: influence of structure

We have described the salient features of radioactive decay and particle emission; it now remains to discuss the relation between the rate of decay and the disintegration energy available in the transition. If we take the empirical approach to this subject and tabulate the maximum electron energies against the corresponding half-lives we see at once that there is a strong correlation between great maximum energy and short half-life. If we follow the procedure first used by Sargent and plot the logarithm of half-life against the logarithm of maximum energy we obtain clear indications that several groupings of radioactive elements exist and that within each group there is a relatively simple relation between maximum energy and half-life. In fact, if we set  $t_H E^5 = K$ , where  $t_H$  is the half-life,  $E$  the maximum energy, and  $K$  a quantity which is dependent on the atomic number and the particular grouping to which the radioactive element belongs, we obtain a very good description of the relation.

This empirical study is illuminated by viewing it in the light of the theory of beta-ray decay. On this theory we consider the parent nucleus and the daughter nucleus and inquire as to the probability of the passage from one to the other. The theory must include the following considerations: First, there is the actual process of change from neutron to proton, electron, and neutrino, which we designate  $G_F^2$  (the subscript  $F$  is for Fermi). Second, there is the term  $S$  which is concerned with the similarity or otherwise of initial and final structures. For some transitions the structural arrangements are such that the skids are greased; these are called "allowed" transitions. For others there is great structural change and such transitions are "forbidden." Third, the electron is influenced by the coulomb field in a manner dependent on its energy  $E$  and the nuclear charge  $Z$ . This we denote by  $F(Z, E)$ . Finally, the electron and neutrino share their energy so that if the electron gets an amount  $E$  and the maximum available is  $E_m$  the chance of its getting that amount is governed by



a term of the form  $E^2(E_m - E)^2$ . The grand total of all these factors gives an equation

$$W_E^{E+dE} dE = G_F^2 S F(Z, E) E^2 (E_m - E)^2 dE \quad (5)$$

where  $W_E^{E+dE}$  is the probability that an electron having energy between  $E$  and  $E + dE$  will be emitted in 1 second.

We can obtain much valuable information from equation 5. In the first place we can see the reason for the relation between half-life and maximum energy. If we consider the *total* probability that an electron is emitted in 1 second no matter what its energy, we are considering the quantity  $\lambda$  of equation 4. This total probability can be obtained by integrating equation 5 from 0 to  $E_m$ , and since

$$\int_0^{E_m} [E^2 (E_m - E)^2] dE = \frac{E_m^5}{30}$$

we obtain

$$\lambda = K_1 E^5$$

where  $K_1$  is approximately a constant, or, because  $t_H = \ln 2/\lambda$ ,

$$t_H E^5 = K \quad (6)$$

where  $K$  is again an approximate constant. This is the relation found empirically.

It is now of interest to study the nature of  $K$ . This can be seen from equation 5 to involve  $G_F^2$ , the term describing the transition from neutron to proton, a term which will presumably be the same for all such radioactive changes; and  $S$ , the term which we said involved the structure of the initial and final nuclei. Now the term "structure" applied to the atom requires care. It is one of the tenets of quantum mechanics that the actual detailed appearance of the atom cannot be ascertained, so that, although the atom clearly has structure, it is useless to attempt to describe it, as it were, photographically. Theory has shown that the quantities most concerned with structure are angular momentum and "spin," and it is in terms of these that we must discuss the effect of structure in nuclei. For example, an atom can be in a state in which its energy is so distributed that there is no net angular momentum. The distribution of the function which gives the chance of finding an electron at any place is then spherically symmetrical. Atoms with varying amounts of angular momentum have varying kinds of symmetry ranging from cigar shape to quite complicated shapes. The term  $S$  will thus be concerned with the angular



momenta of the initial and final nuclei. It will also depend on the actual size of the nuclei. If these two factors can be considered the term in  $S$  can be useful in predicting nuclear transitions.

We can dismiss the effect of nuclear size fairly shortly. Nuclear size changes relatively little from nucleus to nucleus so that the expected effect of this quantity would be to cause a gradual difference between elements as the atomic weight became greater. This is probably the reason why, even in the same grouping, elements of high and low atomic number do not have the same numerical value for  $K$  in equation 6. The effect of shape, or of angular momentum, is much more important. Atomic transitions are profoundly affected by the angular momenta of the two states involved in the transition. A good example of this is given by ortho- and para-hydrogen. The difference between these two forms of hydrogen is in the directions of the "spins" of the two protons forming the nuclei of the molecule. In para-hydrogen the spins of the two are opposite so that their contribution to the total angular momentum is zero; in ortho-hydrogen the two are in the same direction so that the total nuclear angular momentum is (in conventional units) unity. This apparently small difference is sufficient to render the transition from one to the other completely forbidden; the transition can take place only if a third body is present to introduce some perturbation which renders the change possible.

It is therefore easy to see that a radioactive change which required the passage from one nucleus to another with very different angular momentum would be "forbidden" and that therefore  $S$  would be abnormally small. The calculation of such transition probabilities is very difficult, but numerous experimental cases bear out the truth of this analysis. One example can be given. It has been known for a long time that potassium is radioactive. It has been shown that the isotope responsible is  $K^{40}$ , and that the electron energy has the maximum value of 1.4 Mev. We can compare this with  $Si^{31}$  having a maximum energy of 1.8 Mev and a half-life of 2.8 hours. Applying equation 6 we expect  $K^{40}$  to have a half-life of about 10 hours. This short time would preclude its existence as a nearly stable isotope, which fact requires a half-life of more than  $10^8$  years. The transition from  $K^{40}$  to  $Ca^{40}$  thus is highly "forbidden," and this means that a great difference of angular momentum between the two nuclei must exist. Theory suggests that such a difference as three or four conventional units would be needed to explain the relative stability of  $K^{40}$ . Zacharias has shown that the spin of  $K^{40}$  is 4, whereas that of  $Ca^{40}$  was known to be zero. The theory is thus nicely verified. The



explanation of the various groupings is thus that transitions are either "allowed," with no change of angular momentum, or "forbidden," with a change of one or more units. Within each grouping equation 6 holds.

In the table below we give some sample elements and the values for  $t_H E^5$ . For positron emission the observed upper limit is increased by 1 Mev to allow the formation of the positron. The figures are given in Mev to the fifth power times minutes.

ELEMENT	$t_H E^5$	GROUPING
C <sup>10</sup>	240	1
C <sup>11</sup>	590	1
N <sup>13</sup>	505	1
B <sup>12</sup>	91	1
O <sup>15</sup>	290	1
F <sup>20</sup>	630	1
Na <sup>24</sup>	4,750	2
SI <sup>31</sup>	3,200	2
P <sup>32</sup>	285,000	3
Cl <sup>38</sup>	117,000	3
V <sup>52</sup>	142	1
Mn <sup>56</sup>	37,000	3
As <sup>78</sup>	350	1
Br <sup>78</sup>	2,500	2
Ag <sup>110</sup>	33	1
Ag <sup>108</sup>	395	2 or 1
Np <sup>239</sup>	770	2

The groupings can be seen reasonably clearly, although at the same time there is no exact constancy or well-marked trend. It can be seen that the product tends to become less as the atomic number increases, as would be expected from the influence of the increase in nuclear size, which tends to increase the transition probability. More accurate grouping also should include the quantity  $F(Z, E)$ .

The utility of this rule is more apparent when it is applied to predict maximum energies. Thus, for example, Ne<sup>23</sup>, which has a half-life of 40 seconds, can be assumed to be in the first grouping with a value of  $K$  close to that of F<sup>20</sup>, say 600. The value of  $E^5$  is then  $600/0.66$  or 900, and  $E$  should be 3.9 Mev. Actually it is 4.1 Mev as found by absorption. The same reasoning predicts a value of 3.3 Mev for the positrons from Na<sup>21</sup>, whose maximum energy had not been measured at the time of writing. It is possible that by the time of publication this will have been accomplished, and the reader may be able to judge the value of the rule for himself.



## Nuclear isomerism

While we are considering energy relationships it is in place to describe the phenomenon of *nuclear isomerism*, sometimes referred to as gamma-ray decay. It is the case where a single radioactive isotope could have more than one decay period and yet decay to the same element. A little earlier in this chapter we said that a nucleus would not "hold" excitation energy if it could release it as radiation; the time of excitation is of the order of  $10^{-13}$  second. We also, however, men-

tioned the possibility of exceptions to this rule. The exceptional cases are those in which the amount of excitation energy is small and also where the change to the ground state involves a considerable change of angular momentum. Now if we consider a *parent* nucleus which is formed in an excited state of such a sort, it might remain in a state of excitation long enough for it to decay before it lost its excitation energy, in which event the excess energy would be added to the energy normally available for the beta ray and an abnormally fast electron would emerge. This idea can be seen more easily from Fig. 7, in which an energy-level diagram similar to Fig. 3 is shown for two

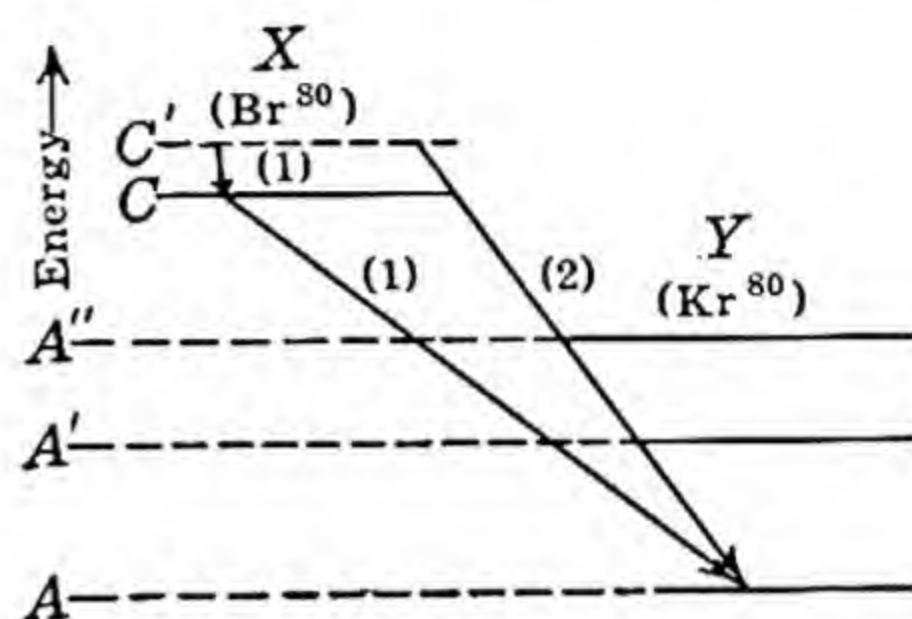


FIG. 7. Nuclear isomerism. An unstable element  $X$  has, in addition to the ground state  $C$ , a metastable state  $C'$  whose angular momentum differs greatly from that of  $C$  so that the transition  $C'$  to  $C$  takes place about as rapidly as the direct decay from  $C'$  to  $A$ . Then the element  $X$  partly decays from  $C'$  as in (2) and partly by the double change, first to  $C$  and then to  $A$  as in (1).

hypothetical elements  $X$  and  $Y$ . The element  $X$  has an excited state  $C'$  a little above the ground state  $C$ ; this level is metastable, probably on account of a great difference of angular momentum between it and the ground state. Now, if it takes a long time for the transition from  $C'$  to  $C$  to occur, there will be time for the element  $X$  to decay while still in the excited state and the energy of excitation  $CC'$  will be given to the resulting beta ray. Now the decay by method (1) of the diagram, by first passage to the ground state and then the final transition, depends on the time it takes for the passage to the ground state, and this is not connected with the ordinary beta decay period. We thus expect that there will be two rates of decay: the ordinary beta decay, as indicated in the diagram by (2), and the double transition, by route (1). It can be seen that this phenomenon of nuclear isomer-



ism, many examples of which are now known, is likely to be complicated also by the transition in either case to excited states. The process of sorting complex beta-ray spectra is still going on. One example of nuclear isomerism which is of interest in applied radioactivity is  $\text{Br}^{80}$ , which decays to  $\text{Kr}^{80}$  with two half-value periods: 18 minutes by direct beta decay, as in route (2) of the diagram; and 4.4 hours by the double transition of route (1). A question has arisen whether the excited nucleus in state  $C'$  would modify the chemical reactions of the atom itself. This is an important question to answer as it affects the whole validity of the tracer method.

Segré and Helmholtz have given a convenient formula which can be applied to numerical estimates of what the likelihood is for nuclear isomerism. If a gamma-ray transition involves an energy  $E$  (Mev) and a change of angular momentum of  $l$  in an element of atomic number  $A$ , an estimate of the decay constant  $\lambda$  is given by

$$\log_{10} \lambda = 20.3 - 2 \log_{10} [1 \cdot 3 \cdot 5 \cdots (2l - 1)] - (2l + 1)(1.30 - \log_{10} E) - 2l(0.84 - \frac{1}{3} \log A) \quad (7)$$

This rather clumsy-looking formula is very convenient to use and most instructive. If, say,  $E$  is 1 Mev,  $A$  is 50, and  $l$  is 1, the value found for  $\lambda$  is  $7.9 \times 10^{15}$  inverse seconds. However, if  $E$  is 0.1 Mev,  $A$  is 100, and  $l$  is 4, the completely different result  $\lambda = 1.4 \times 10^{-6}$  inverse second is obtained. The two corresponding half-lives are  $8.7 \times 10^{-17}$  second and 5.7 days. Nuclear isomerism thus represents those cases where nuclear structure requires a large change of  $l$  for the emission of a gamma ray with attendant long half-life. Hence it is also referred to as gamma-ray decay.

### Emission of gamma radiation

Although the emission of gamma radiation has been mentioned as a natural consequence of the possibility of decay to an excited state of the product nucleus, a little elaboration will do no harm. If one looks through the list of radioactive elements and their properties given in Appendix 2 it can be seen that about a third of the elements are simple, in the sense that they emit only beta rays; the remainder emit gamma rays in addition. This emission of gamma rays is a great help in many counting experiments as their presence permits the detection of the radioactive material through great thicknesses of material. The amount of gamma radiation emitted depends on the relative numbers of transitions to excited states. Thus no general rule can be stated as



to which elements will give intense gamma radiations, and it is necessary to consult tables to obtain information on this point.

### Internal conversion

In some cases a fraction of the transitions from an excited state to a lower state is achieved by conveying energy to one of the electrons in the extranuclear structure. This is called *internal conversion* and is of great theoretical interest as it is a process which can be rather thoroughly treated theoretically. The effect of internal conversion is to inject sharp lines onto the beta-ray spectrum. In some cases of gamma-ray decay there is no continuous beta-ray spectrum but only the internal conversion line. The lines are of energy determined by the gamma-ray energy less the electronic binding energy.

It is possible for a sufficiently energetic transition to produce a positron-electron pair near the nucleus. This is a second category of internal conversion, called *internal pair formation*.

### Positron emission and K-electron capture

We have made the point that the process of radioactivity, the passage from an unstable nucleus to a more stable one, does not imply that the adaptation always requires the change of a neutron into a proton, electron, and a neutrino; the reverse process of the transition of a proton into a neutron, a positron, and a neutrino can also take place. If this occurs the detected charged particle is now a positron, and, as one or two features of positron emission differ from electron emission, they should now be considered. The first difference has already been mentioned: the fact that a positron is really a "hole" in the states of negative kinetic energy requires that at least 1 Mev of energy be supplied to eject an electron from a place in these states of negative kinetic energy and so create a positron. If there is less than 1 Mev of available energy due to the difference of mass between the initial atom and the daughter atom, the emission of a positron cannot take place, and in any event the kinetic energy possessed by the positron for a given amount of disintegration energy is always less than that of an electron by this amount of 1 Mev. Thus  $N^{13}$  emits positrons with a maximum energy of 1.20 Mev, while the difference in mass between  $N^{13}$  and the product  $C^{13}$  is 2.22 Mev.

The reader will have natural curiosity as to the events expected if two atoms are found in which the difference of mass is less than 1 Mev, and such that the more stable requires the emission of a positron from the less. Such atoms do occur, and the passage to stability is



effected by the absorption of an electron from the outer atom, again one of the cases in which we become conscious of the existence of the ordinary electronic structure. In any atom the innermost shell of electrons, the  $K$  shell, possesses no angular momentum, and therefore the function which describes the probability of finding one of these electrons has a definite value inside the nucleus. One is not surprised that this fact makes it possible for an unstable nucleus to have a tendency to absorb this electron permanently and so gain stability. This process, known as  $K$ -electron capture, has several interesting features.

They can be understood from a definite example. The nucleus  $V^{47}$  has been shown by Walke, Williams, and Evans to be of this type; it is radioactive by  $K$ -electron capture to form  $Ti^{47}$ . The half-life is 600 days. There is not sufficient energy to cause positron emission, and so an electron of the  $K$ -shell is absorbed by the nucleus. This now produces  $Ti^{47}$ , *with an electron missing in the  $K$ -shell*, which missing electron must be replaced to render the atom neutral. The filling of this shell causes the emission of the x-rays characteristic of the  $K$ -shell of *titanium*, not vanadium, and by the presence of this x-radiation the process of  $K$ -electron capture can be detected; by showing the radiation to be characteristic of titanium and not vanadium the explanation of the process is shown to be correct. Notice that to detect  $K$ -electron capture we need counting technique which will respond to rather soft x-radiation. It may happen that enough energy is available to cause the absorption of the  $K$ -shell electron and the emission of gamma radiation also, as in  $Be^7$ , which goes to  $Li^7$  with the absorption of an electron and also the emission of a 0.45-Mev gamma ray. Notice that the energy of the gamma ray is always rather small, generally below 1 Mev. Where this gamma radiation is also emitted the detection is easier as ordinary counters are satisfactory.  $K$ -capture becomes very important in the heavier elements where the electron density near the nucleus is high.

Positron emission is always accompanied by a by-product in the form of annihilation radiation. The positrons have a transient existence, and, especially in the presence of absorbing material of high atomic number such as lead, there is a considerable production of 0.5-Mev gamma radiation due to the "annihilation" of the positron, or rather the filling of the "hole" in the states of negative kinetic energy, which appears to be a positive particle. This was explained more fully in Chapter 2. The result is that there is always the appearance of gamma radiation from positron emitters, but great care must be taken if the radioactivity is detected by means of this radiation, as the



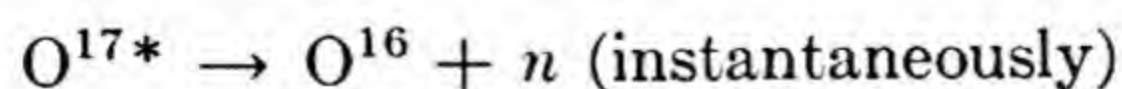
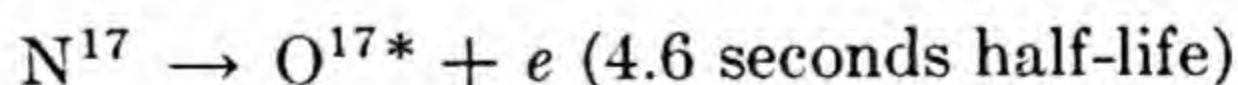
amount varies with the nature of the material around the apparatus, thus giving erratic readings. The annihilation radiation is not generated in the primary radioactive substance, but as a secondary product in absorbing material near by.

### "Parent" and "daughter" elements ✓

We have so far described the fact of radioactivity as the process of reversion to type. Only a limited number of processes is available for this readjustment, and it is therefore not surprising if the final adjustment does not take place in one radioactive change. Many such atoms are known, as for example the decay of  $\text{Co}^{55}$  into  $\text{Fe}^{55}$  by the emission of a positron, followed by the change of  $\text{Fe}^{55}$  into  $\text{Mn}^{55}$  by  $K$ -capture. The  $\text{Mn}^{55}$  is finally stable. The second radioactive element is called the "daughter" of the first "parent" element. The discovery of the growth of one element from another is often of the greatest value as a means of identification.

### Delayed neutrons

Perhaps one of the most interesting parent-daughter processes is that by which delayed neutrons are produced. These were originally discovered in connection with uranium fission, and their importance in the control of a chain reacting pile is described in Chapter 11. An example of the process which occurs is afforded by the nucleus  $\text{N}^{17}$ . This property of the  $\text{N}^{17}$  nucleus was discovered by Alvarez. This can be produced by bombarding  $\text{C}^{14}$  by alpha particles according to the reaction  $\text{C}^{14}(\alpha p)\text{N}^{17}$ . The  $\text{N}^{17}$  can readily decay to  $\text{O}^{17}$  which is stable. However, there must be a strong structural difference between the  $\text{N}^{17}$  and  $\text{O}^{17}$  in its ground state, for the  $\text{N}^{17}$  prefers to decay to  $\text{O}^{17}$  in a state of high excitation. This highly excited oxygen is capable of splitting off a neutron to form  $\text{O}^{16}$ , a process which occurs essentially instantly. The daughter element  $\text{O}^{17*}$ , where the star indicates excitation, thus emits a neutron but the rate is controlled by the half-life of the parent element which is 4.6 seconds. The combined process is then



### Alpha-particle emission

It has been stressed that radioactivity is the passage from an unstable to a stable nucleus. In the foregoing it has been assumed that



the adaptation is by the change of a neutron into a proton, or vice versa, but there is no real reason why this should be the only method of adaptation. Any method which permits stability to be reached will be expected to take place, but not all such methods will give rise to radioactivity, which is a process which occupies time. We have just seen that the emission of a neutron is possible, but it is instantaneous. The emission of a proton might not be instantaneous because the penetration of the barrier might act to diminish the probability of

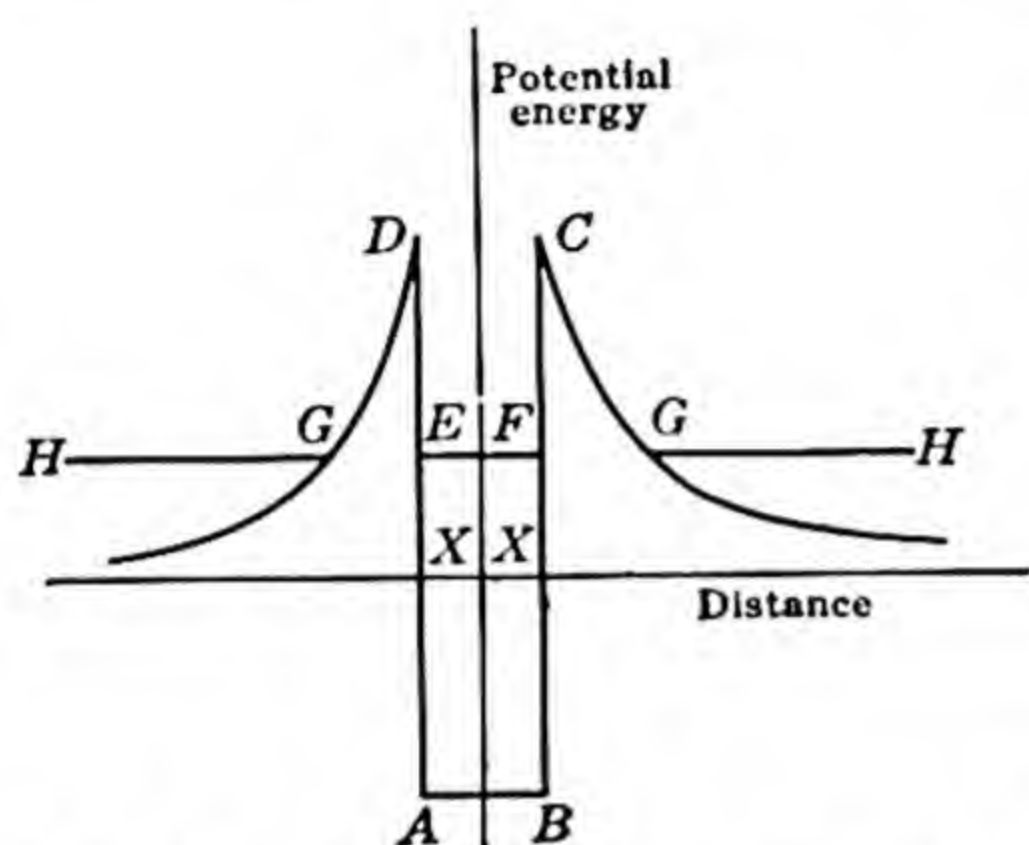


FIG. 8. Alpha-particle emission. The alpha particle has enough excess energy  $EX$  to escape but is held back by the potential barrier. It therefore continues to move from  $E$  to  $F$  inside the well, but as the barrier has a finite thickness the particle has a finite time to escape at each collision with  $E$  or  $F$ . In time it will do so, its half-life being less, the greater the available energy  $EX$ .

emergence. Such barriers are not very high and the time to decay might still be short. However, the barriers opposing alpha-particle emission are twice as high, and in the case of very heavy nuclei with large nuclear charge it is possible that the time for escape of a relatively energetic particle might be long.

This is found to be the case. Many elements, notably uranium and radium, emit alpha particles with long half-lives. These are  $10^9$  and 1830 years, respectively. Actually both these elements are unstable in a totally different sense from beta emitters. The nucleus can almost be thought of as already split apart, with an alpha particle ready to emerge, but the barrier holds back the alpha particle until the penetra-

tion phenomenon permits it to leak out. We wish to make the point that any form of adaptation to produce stability can be thought of in a general way as radioactivity; alpha-particle emission is more favorable for long-lived processes of adaptation.

The details of the process are usually explained by the type of diagram in Fig. 8. This is similar to the diagrams of transmutation processes shown in the previous chapter. The alpha particle is thought of as moving under the influence of the rest of the nucleus, which will be the daughter nucleus after the separation has taken place. It is supposed to have enough energy of excitation so that it could escape, a fact represented on the diagram by a positive value for the line  $EF$



which denotes the level occupied by the alpha particle. The existence of the potential barrier, which is due to the superposition of the coulomb repulsion on the attractive nuclear field, renders the escape of the alpha particle possible, but subject to being hampered by the necessity of penetrating the barrier: in more accurate language, the alpha particle has, at each collision it makes with the walls at  $E$  or  $F$ , a finite chance of penetration. The greater this chance, the fewer transits of the distance  $EF$  will be necessary before the alpha particle escapes. This chance depends markedly on the height and thickness of the barrier to be penetrated, and these in turn depend on the distances  $EG$  and  $ED$  or finally on the degree of excitation of the alpha particle if the shape of the barrier is, in general, the same for different cases. We therefore predict that if fast alpha particles are observed the half-life of the emitting element will be short, and if the alpha particles are slow the half-life will be very much longer. This explains the rule found empirically by Geiger and Nuttall, that the logarithm of the half-life is proportional to the logarithm of the energy of emission of the alpha particles. It is not exact, and this deviation from exactness is laid to the variation in the potential barriers from one radioactive element to another. Alpha-particle emission is thus observed virtually only among very heavy elements where the potential barriers are high and also the increased importance of the coulomb forces in the nuclei renders elements unstable with respect to alpha-particle emission.

### Radioactive series

The passage to stability is not always achieved in one change. If the way in which an element is formed is unrelated to its own structure, as happens in nuclear fission, there can result a series of radioactive processes which ultimately achieve stability. The most striking examples of such series are found where alpha-particle emission can take place. In the region of the elements where alpha-particle decay takes place, there is a great excess of neutrons over protons. The emission of an alpha particle, which takes out two protons with only two neutrons attached to them, tends to increase this excess, and it may well happen that the excess is too great for stability, which accordingly gives rise to ordinary electron emission. This means that the alpha-particle emission is accompanied by beta-ray emission from the daughter element, and we have a chain of radioactive decay. Three such chains have been known for a long time: the thorium,



uranium, and actinium series. The feature of these series is the approximate alternation of alpha-particle emission with beta-ray emission. They may be explained roughly as due to the tendency for the highly charged nuclei to eject alpha particles, followed by a tendency to diminish the resulting neutron excess by beta decay.

The subject of natural radioactivity is covered quite satisfactorily in several standard works, to which we refer the reader. As far as the use of naturally radioactive elements in tracer work and other applications is concerned, they may be treated as being the same as artificial products.

To summarize this chapter we may repeat that radioactivity is the passage from an unstable nucleus to a more stable, the change generally being accompanied by the emission of a charged particle which has enough energy to ionize. The commonest process of adaptation is the change of a neutron into a proton, electron, and neutrino; or a proton into a neutron, positron, and neutrino; the rate of the change depends on the available energy as determined by the relation  $E^5 \times$  (half-life) is roughly constant. The energy is divided *on the average* nearly equally between the electron or positron and the neutrino, resulting in a bell-shaped distribution curve whose maximum value is the "disintegration energy." The rate of decay is proportional to the number of atoms ready to decay, giving a logarithmic decay curve. The existence of excited states in the daughter nucleus can give rise to the emission of gamma radiation, which modifies the distribution curve to a superposition of several curves and favors the emission of electrons of less energy. If a metastable state exists in the parent element, two decay periods are possible for the same radioactive change—the phenomenon of nuclear isomerism. Positron emission differs from electron emission in requiring the extra energy of 1 Mev to create the positron and also in the accompaniment of "annihilation radiation." When there is insufficient energy to effect the creation of a positron, an electron is captured instead, a process known as *K*-electron capture. Alpha-particle emission occurs among the heavier elements where there are high potential barriers and considerable coulomb forces; it is generally accompanied by subsequent beta emission giving rise to a radioactive series.

We have now covered the necessary account of the nature of nuclear physics sufficiently to make its applications intelligible; in the next chapter we shall consider the manufacture and counting of radioactive elements in some detail.



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## 7. Technique in Artificial Radioactivity

This chapter will frankly be written for the individual who is interested in making actual measurements that involve artificial radioactivity. If we were considerate we would advise the general reader to pass on to more rewarding chapters, but as he has taken the trouble to obtain this book we do not see why we should not make the claim that the material in this chapter is as interesting as the remainder and let him prove us to be in the wrong after perusal. The physicist is often accused of being rather aloof, of regarding the applications of his subject as beneath his consideration once he has mastered it; and it may seem to some of our readers that we have in a sense been "holding out" on them until now, for we have not faced any practical problems as yet, and the question may be arising whether we ever intend to do so. This chapter is the answer.

We may begin with the problem of observation of various elements since it is usually this problem that the actual worker has to solve while he is waiting for the physicist to manufacture the sources for his experiments. The question to be answered will always appertain to a particular problem and cannot be answered without thought applied to that problem itself. For example, one might be interested in a problem requiring radioactive sulphur, and a glance at some tables would show that  $S^{35}$  has a half-life of 88 days and emits a beta ray. This is a sufficiently long half-life to make it plausible that  $S^{35}$  could be used in experiments in animals and followed during their lifetime by counters outside the animal. Actually the beta ray has a maximum energy of only 0.1 Mev; no gamma ray is emitted; and the sulphur would be undetectable through even the skin of the animal. The experiment would therefore fail.

### General considerations in counting

With this example it may appear that the problem of determining whether a piece of research is to be attempted is one for the expert



alone to decide. This is only partly true. A great deal of the problem of counting is related to the question of whether the radiation from the radioactive substance can enter the counting mechanism or not. This means that *absorption* is of prime importance in deciding the feasibility of an experiment. Now in the second chapter we described the various types of radiation, and in the third the various types of counting technique. It generally happens that detection is by means of the effects of beta radiation, and therefore it is in place to consider the absorption of beta radiation first. We made the point in the second chapter that, although a beta ray of a definite energy had a certain maximum range, its path through matter was devious and one could not be sure that the thickness of absorber which would stop the radiation from reaching a counter would represent the thickness corresponding to the energy of the particular electron. This is a real limitation in making measurements of beta-ray energies by absorption, but it is not particularly annoying if one is merely interested in making sure that counting methods will work in a given experiment. In practice one can allot to a beta ray of a given energy a definite thickness of material which it can penetrate. One must be ready to find that this is an average value, not accurately followed every time, but a perfectly good guide. The thickness of material depends on the substance used to act as absorber, but not markedly so if one expresses the absorption in terms of *mass per square centimeter* instead of actual thickness.

By and large the absorption depends on the numbers of electrons passed in the material, and in a square centimeter of any material having the same weight there are about the same number of electrons; thus the thickness of gold which will just stop a 1-Mev electron is roughly 0.5 gram per square centimeter, whereas the same electron would have been stopped by 0.4-gram per square centimeter thickness of aluminum. In point of fact the thickness (mass per square centimeter) of any material which will stop an electron of given energy depends on the ratio of the nuclear mass to the nuclear charge; the greater this ratio the less effective the material is, and therefore the greater the thickness which must be interposed to be correspondingly effective.

It may happen that the idea of measuring the thickness of an absorber in mass per unit area is unfamiliar to the reader. If this is so we might mention that it is common practice in all kinds of nuclear absorption work to use this method of expression, partly because very thin films can be measured more accurately this way, and partly be-



cause it comes close to measuring the really effective quantity; if one measured the number of electrons per unit area it would be still better, but this is rather too far from ordinary practical measurement. To take a definite example, we could consider a thickness of 1 mm of aluminum, which is a very average absorption thickness for beta-ray experiments. This would have a volume of 0.1 cc for every square centimeter of its area; and, as its density is 2.6, the mass per unit area would be 0.26 gram. We can thus see from the figure quoted above that to stop a 1-Mev electron would require a 1.52-mm thickness of aluminum. The same reasoning leads to a thickness of 0.26 mm for gold. As a very rough guide we can give the rule:  $t \times d = 0.54E_m - 0.16$ . Here  $t$  is the *thickness measured in centimeters*,  $d$  is the density in grams per cubic centimeters, and  $E_m$  is the maximum energy of the electron in Mev. This rule must be considered a rough guide only, but with that proviso it applies to any substance used as absorber. We shall refer to it as the *rough beta-ray formula*.

We can now try out the rule for one or two examples and see what else we need to know. Let us suppose that we wish to count radio-phosphorus,  $P^{32}$ , with its maximum energy of 1.7 Mev. Suppose also that we have available a counter with a glass wall 1 mm thick and a copper electrode 0.25 mm thick. How effective will this be, and is much to be gained by an elaborately constructed thin-walled counter or electroscope? The density of glass is 2.5 gram per cubic centimeter; of copper, 8.9 gram per cubic centimeter; using the rough beta-ray formula we find that this counter will stop all electrons of less energy than 1.2 Mev. In obtaining this result we add the separate products of thickness and density to get the left-hand side of the rough beta-ray equation. A glance at tables will show that  $P^{32}$  does not emit any gamma radiation, and we can therefore plot a rough distribution curve for the emitted electrons by drawing a bell-shaped curve with the maximum at the one-third-limiting energy of 0.55 Mev: this shows roughly that one-sixth the area under the curve or one-sixth the total number of electrons emitted has energy greater than the value of 1.2 Mev corresponding to the least energy which will suffice to enter the counter. Thus, though there is no doubt that the counter will readily detect the electrons from  $P^{32}$ , it will be rather inefficient. It would have been better to use thinner glass and thinner copper; the rough beta-ray formula shows that halving the thicknesses of both would permit electrons of 0.75 Mev to register, and this would count 40 per cent of the total electrons emitted.



All further improvements are not very rewarding as it gets increasingly difficult to diminish thickness and the counter is already sensitive to the majority of the electrons emitted. To save the reader a little trouble we might say here that an ordinary counter, constructed without special precautions, will permit beta rays of energies between 0.6 and 0.9 Mev to register. Thin-walled counters can be made which will count as low as 0.05 Mev; below this it is necessary to use electroscopes or actually put the material in the counter itself.

Before we consider these more elaborate techniques we might apply the rough formula above to another substance,  $\text{Ca}^{45}$ , the 180-day calcium isotope. This has a maximum energy of 0.25 Mev, and as it has a long half-life, and is difficult to prepare in large amounts, its detection is likely to be a borderline matter. The counter should therefore be of the thin-walled variety, and then a great advantage will be gained over the use of an ordinary counter. In Appendix 4 we present a discussion of the absorption of beta rays which will enable the appropriate counting technique to be decided. In self-defense we should say here that we do not claim that all the figures given are accurate; they are intended to serve as a guide, not as a reference for research in nuclear physics.

The reader who may be intending to ask for a grant to buy apparatus may like to know why sometimes counters are used and sometimes electroscopes. Both have their advocates; a well-equipped institution should own both. The electroscope is probably the more universally useful, and a determined experimenter can adapt an electroscope to fulfill any function that can be satisfied by a counter except the registering of coincidences. For this reason the counter is used most commonly in cosmic-ray work where whole trays of counters working together are commonplace. The Geiger counter is, on the surface, the most sensitive detecting agent since it unquestionably detects a single fast particle, and no more can be asked of it; this fact, however, is not all, as a counter also has inevitably a background count, of the order of 2 per minute per square centimeter of surface of the electrode, and so it is not really practicable to detect less than half the background. This is a serious limitation on an apparently perfect detector, as it requires a source capable of producing about 10 impulses per minute to detect. The electroscope, supposing that the insulation is perfect and that it is shielded carefully, can have a very slow rate of leak; for example, a Lauritsen electroscope has a rate of leak of about 5 divisions per hour, while a source capable of producing 10 pulses per minute in a Geiger counter will add 2 more



divisions per hour. The source that can be readily detected by a Geiger counter can thus also be detected by a Lauritsen electroscope. The main difference is that it would take two hours to determine the rate of leak of the electroscope, whereas the Geiger counter would have yielded complete information in a few minutes. For rapid work with weak sources the Geiger counter is to be preferred. For ordinary, run-of-the-mill counting of easy elements there is not much choice. The electroscope comes into its own when *slow electrons* have to be counted.

The way in which an electron ionizes as its speed is reduced is of great interest. We give below a short table relating the number of ions produced in a 1-cm path in air by electrons of various energies. Notice that the less the speed the greater the ionization; in fact, a simple relation holds between the *velocity* of the fast electron and the ionization per centimeter. If this ionization is called  $I$ , then  $Iv^2$  is a constant,  $v$  being the velocity of the electron. This relation is a little troublesome to use as the velocity of an electron of given energy must be calculated by allowing for the change of mass with velocity, and the table is a more convenient way of seeing what happens.

TABLE 1

ENERGY OF ELECTRON (Mev)	IONIZATION PRODUCED IN 1 CM OF AIR (ION PAIRS)
0.05	250
0.10	175
0.20	96
0.30	76
0.50	60
1.00	53

The difference between a counter and an electroscope is that the counter is a "trigger-action" instrument, which records large and small ionization in its sensitive volume equally well, whereas the electroscope records the ionization, giving large readings for large ionization. In the usual Lauritsen electroscope there is a path length of about 7 cm; and an electron of 0.1 Mev, which will just traverse the air in the ionization chamber, will produce 1400 ion pairs, whereas a fast electron of, say, 1 Mev will produce only 350 ion pairs. It is clear that the electroscope is operating more favorably in detecting slow electrons than fast, and this, coupled with the fact that there is no need for a thick case to hold a vacuum as in a Geiger counter which would prevent the entry of slow electrons, makes the electroscope very suitable for detecting low-energy electrons.



Before we consider the counting of gamma radiation it is of interest to consider one or two consequences of the ionization relation given above. It may be desired to employ electroscopes technique to count fast electrons which produce relatively little ionization. The sensitivity can then be greatly increased by using an ionization chamber at several atmospheres' pressure. In many respects this is the best way of detecting electrons. The use of high pressure increases the number of atoms of air in the path of the fast electrons on their way through the ionization chamber and so increases the number of ions formed.

To give an example: it would be possible to obtain the same number, 1400, of ions with the same ionization chamber traversed by 1-Mev electrons as was formed by slow electrons; this would be achieved by filling the ionization chamber to 4 atmospheres. This type of ionization chamber has the advantage that it can be made to be recording and is not subject to variations in sensitivity if the pressure is kept the same. It is also common practice to fill the ionization chamber with some heavy vapor such as  $\text{SO}_2$  or methyl iodide, which fulfills the same purpose.

The question of constant sensitivity is important. An electroscopes is generally well protected from mechanical disturbance and will always have the same properties. This is also true of a Geiger counter if it is carefully sealed and is constructed to have a good "plateau" so that extraordinary precautions to ensure constancy of the voltage supply are not required. Nevertheless, the recording equipment is more elaborate, and unless the operator is sure of his business it is quite possible that some factor (e.g., the bias on the thyatron recorder) will vary and this will introduce a varying sensitivity. Therefore it is easier to secure foolproof constancy of operation with an electroscopes than with a counter, and it is especially preferable to follow electroscopes technique for work where long-lived elements are concerned and comparison has to be made between readings taken several days apart. With these considerations in mind the reader can decide for himself which method of observation to adopt.

### **Gamma-ray counting**

The counting of gamma radiation is the next matter to consider. Gamma rays are not counted directly; it is the secondary electrons that they produce that cause the ionization necessary to be detected. This fact should be considered carefully. The radiation has various methods of interacting with electrons (or any charged particle), and these produce various effects. For instance the photoelectric effect



means that the whole energy of the gamma-ray quantum is given to an electron in one occurrence; the formation of a pair of electrons also uses up the whole gamma-ray energy; on the other hand the process of Compton recoil, considered in the next section, imparts only a fraction to the electron. As far as counting the gamma ray is concerned we need mostly to note that the radiation produces secondary electrons which have somewhere near the energy of the incident gamma radiation, and that it is these electrons which must be detected.

It is still true that absorption plays a fundamental part in the design of a detecting apparatus, but in a rather different way because the absorber has two effects: the first is the usual stopping of the electrons; the second is the actual formation of the fast secondaries. The second effect introduces important differences. Electromagnetic radiation will interact only with charged particles, so that unless some charged particles are encountered there will be no effect on the gamma radiation. As soon as material containing electrons is placed in its path the radiation has a chance of interacting with the electrons and losing energy to them; this means that initially the number of secondary electrons increases as the thickness of material interposed increases. What happens as the amount of material is still further increased depends on the energy of the gamma radiation, for, if it is great, the electrons produced will not be absorbed until a considerable thickness of matter is interposed, whereas if it is small the secondary electrons will have rather low energy and will be absorbed by a small thickness of matter. There is thus an increase in the yield of secondary electrons as matter is interposed in the way of the radiation up to a thickness at which the electrons begin to be absorbed. Therefore counters with thick walls are better for gamma-ray counting than thin-walled counters; in fact, there is also an improvement if a heavy element such as gold is used as the electrode of the Geiger counter. Scintillation counters, although they have a high background, are more sensitive to gamma radiation. This is because the electrons produce *light*, which is not rapidly absorbed.

### Absorption of gamma radiation

A word here on the absorption of gamma radiation in general will be useful. The three processes of interaction of radiation with matter—photoelectric effect, pair formation, and Compton recoil—all contribute to the absorption. All three are different in nature. There is, however, the similarity among all three that the moment of interaction



cannot be predicted, only a probability of interaction. This means that gamma radiation is never completely absorbed by any finite thickness of material, for if we have a source emitting many gamma-ray quanta there is always a chance that one of them will not interact with an electron until it has passed the absorption screens interposed, no matter how thick they are.

Notice that any *one* gamma-ray quantum is often completely absorbed; it is a large number from a source that cannot be absorbed *for sure*.

The process of gamma ray absorption follows that of any random absorption process as described on p. 106. The result obtained is

$$\frac{dn}{n} = -\sigma N x$$

where  $\sigma$  is the cross section for absorption and  $N$  is the number of electrons per unit volume. The integral of this is

$$n = n_0 e^{-\sigma N x}$$

or

$$\log_{10} n_0 - \log_{10} n = \frac{\sigma N}{2.3} x = \frac{\tau}{2.3} x$$

The quantity  $\tau = \sigma N$  is called the *absorption coefficient*. The absorption process depends on random interaction with the electrons in the atoms of the material traversed.

With different materials, different absorption coefficients must be used, a fact which is rather confusing and which has led to the practice of writing the absorption coefficient *per electron*, which gives values which can be more readily compared from one material to another. The absorption coefficient per electron is the quantity  $\sigma$  used above and is in reality the cross section per electron. Another practice is to give the mass absorption coefficient, which is the absorption coefficient per gram of material in place of per centimeter of path. The mass absorption coefficient is  $\tau/\rho$  where  $\rho$  is the density in grams per cubic centimeter.

Of the three processes we have mentioned as causing the absorption of gamma radiation, the first two depend markedly on the nature of the element used as the absorber, and the third and most important (in general) is independent of the material but depends only on the number of electrons present for a given energy of the gamma ray.



If we write the absorption coefficient per electron as the sum of three coefficients with self-explanatory symbols,  $\tau_e = \tau_{ph} + \tau_{pa} + \tau_{cr}$ , we can discuss each process separately. Take the photoelectric absorption  $\tau_{ph}$  first. This process requires the transfer of the energy of the "photon" (the gamma-ray quantum) to an electron bound in the atom by a process already familiar in atomic physics. The chance of such an event is great if the energy of the photon is close to the energy of the electron in its orbit, and, as for light elements this never exceeds a few thousand electron volts, the high-energy gamma quantum is unlikely to give up its energy; for heavy elements the energy of an electron in a deep level is of the order of 100,000 electron volts and the chance is much greater. The absorption coefficient for the photoelectric effect  $\tau_{ph}$  is then greatest for heavy elements and relatively low-energy gamma radiation. The coefficient for pair formation  $\tau_{pa}$  will be zero unless the radiation has an energy exceeding 1 Mev for all elements, as pairs cannot be formed unless the energy equivalent to the mass of the pair is available. The theory of pair formation shows that the chance of such an event is greater in the proximity of a heavy element and also greater for energetic radiation; the coefficient therefore increases for increasing atomic weight of the absorber and with the energy of the incident radiation. Finally the third coefficient  $\tau_{cr}$  depends only on the energy of the radiation, decreasing smoothly with energy.

The reader will see that if an absorption coefficient is measured in lead there is a chance that it will correspond to two energies of radiation: one of high energy which is being absorbed mostly by pair formation, and one of low energy which is being absorbed by the photoelectric effect more than any other way. This fact discourages the use of lead as absorbers in experiments to determine the energy of the radiation and leads to the use of aluminum, for which the only effect which is important is the process of Compton recoil, and which leads to a unique value for the deduced energy.

It is not likely that detailed knowledge of gamma-ray absorption will be needed by many readers, but it may happen that a quick determination of the energy of a photon may be required for the purpose of checking the absence of an impurity, or some other reason, so we give here a table of absorption coefficients per electron for the Compton effect alone. If absorbers of light elements such as aluminum are used, these values will be accurate for energies up to 3.5 Mev, which covers most radioactive elements.



ENERGY OF GAMMA RAY (Mev)	ABSORPTION COEFFICIENT PER ELECTRON	HALF-VALUE THICKNESS IN ALUMINUM (cm)
0.5	$2.7 \times 10^{-25}$	3.1
0.8	2.3	3.8
1.0	2.0	4.2
1.2	1.7	4.5
1.5	1.6	5.0
2.0	1.3	5.8
2.5	1.2	6.5
3.0	1.1	7.1
4.0	0.8	8.2

To save a little time in calculation we include also figures for the half-value thickness in aluminum; this thickness is the thickness which will absorb half the radiation on the assumption that only one energy of gamma ray is present. If several energies are involved, as is frequent, the curve for the logarithm of the ionization versus thickness must be decomposed in a manner similar to that about to be described for the logarithmic decay curves found when several radioelements are present simultaneously; to each single line so obtained there will correspond a half-value thickness characteristic of the particular gamma quantum involved.

With this information and the understanding of the general features of detecting electrons, the reader can decide for himself whether any given counting arrangement is satisfactory for measuring gamma rays. It will be noticed that the yield of secondary electrons obtainable is not greatly dependent on the energy of the radiation, for the greater the energy, the thicker the material which will evolve secondaries before they are absorbed.

### Complex decay curves

In the last chapter we described the features of a simple decay curve, and in the above section we have explained that a single gamma energy gives an exponential absorption curve. Both these cease to be simple exponentials when more than one radioactive substance is present in the plotting of decay curves or when more than one gamma quantum energy is present in the plotting of absorption curves. What can be done to sort out the complications which result in such cases? Let us consider the first case of superposed exponentials, namely, decay when several elements having different periods are present. Suppose that we have a target fresh from the cyclotron, for example



radiophosphorus. This is supposed to have a half-life of 14 days, and if we put the target in front of a Lauritsen electroscope and observe the decay of the activity, we would expect that it would remain constant during the first day at any rate. Actually it is almost certain that the rate of decay would show a marked fall within the first 10 minutes and that even after a day or two there would be a deviation from the simple logarithmic diminution. The reason for this deviation is the presence of impurities, of which the two most likely are  $N^{13}$ , of half-life 10 minutes, and  $Na^{24}$ , of half-life 15 hours. The first is due to the presence of carbon, an almost universal contaminant, the second to sodium, the two reactions being  $C^{12}(dn)N^{13}$  and  $Na^{23}(dp)Na^{24}$ . The target thus contains three radioelements in place of one, and each of them decays with its own characteristic half-life. If necessary, the three could very easily be shown separately experimentally. Thus  $N^{13}$  emits positrons, and by using a magnetic field to bend out the electrons the activity of  $N^{13}$  alone could be detected; also  $Na^{24}$  emits hard gamma rays, and by interposing several centimeters of lead as absorber the activity of  $Na^{24}$  alone could be followed; the activity of the phosphorus would then be the difference between the separate effects of the sodium and the nitrogen and that of the total (a little correcting would have to be done for the different sensitivity of detection of gamma radiation and beta radiation).

We can then see that the simple decay curve obtained without any such experimental sorting out would be the sum of three curves, each of which is a simple exponential, following a definite manner of decay. In fact, we could write the equation of the complex curve as

$$N = N_1 + N_2 + N_3 = N_n e^{-\lambda_1 t} + N_{na} e^{-\lambda_2 t} + N_p e^{-\lambda_3 t}$$

where  $N$  is the total number of radioactive atoms present at any time;  $N_1$ ,  $N_2$ , and  $N_3$  are the numbers of atoms of nitrogen, sodium, and phosphorus present at any time;  $N_n$ ,  $N_{na}$ , and  $N_p$  are the initial numbers of nitrogen, sodium, and phosphorus produced by the bombardment; and  $\lambda_1$ ,  $\lambda_2$ , and  $\lambda_3$  are the three decay constants for the three elements. A similar relation holds for the activities. If we write the above relation in terms of logarithms we get

$$\ln N = \ln (N_n e^{-\lambda_1 t} + N_{na} e^{-\lambda_2 t} + N_p e^{-\lambda_3 t})$$

This will not give a straight-line graph for the logarithm of the activity against time. However, if we wait several days we find that a graph so plotted actually straightens out and obeys the relation for the simple decay of radiophosphorus. Looking at the equation above



we see that the two first terms on the right-hand side approach zero in a few days; the first, indeed, does so in a few hours. Then the equation becomes

$$\ln N = \ln (N_p e^{-\lambda_3 t}) = \ln N_p - \lambda_3 t$$

We can therefore use this simple relation to calculate the number of phosphorus atoms at any time or, more directly, the activity due to phosphorus at any time. This can then be deducted from both sides of the composite equation above, leaving an equation involving the activity due to nitrogen and sodium only. In turn the logarithmic plot of this activity straightens out after a few hours and enables the activity due to sodium alone to be deducted. This finally leaves the nitrogen alone.

This kind of "peeling off" is made very easy by plotting on semilogarithmic graph paper. Such paper can be improvised by laying off the units for the ordinates from a slide rule. With such a graph, as indicated in Fig. 1, the line found at long times can be extrapolated back to zero time and the corresponding activity read off directly. This can then be deducted from the composite activity very easily, and a new curve drawn giving only the sodium and nitrogen. The same treatment can now be applied to this curve, the sodium deducted, leaving only the nitrogen. The de-

composition of a complicated curve in this manner is only a matter of minutes. The various lines are shown in Fig. 1. Notice that, if each straight line is carried back to the time corresponding to the conclusion of the bombardment, the yield is given by the value at that point; and also that if the analysis has been carried out carefully there will be agreement between the slopes of the lines and the accepted decay

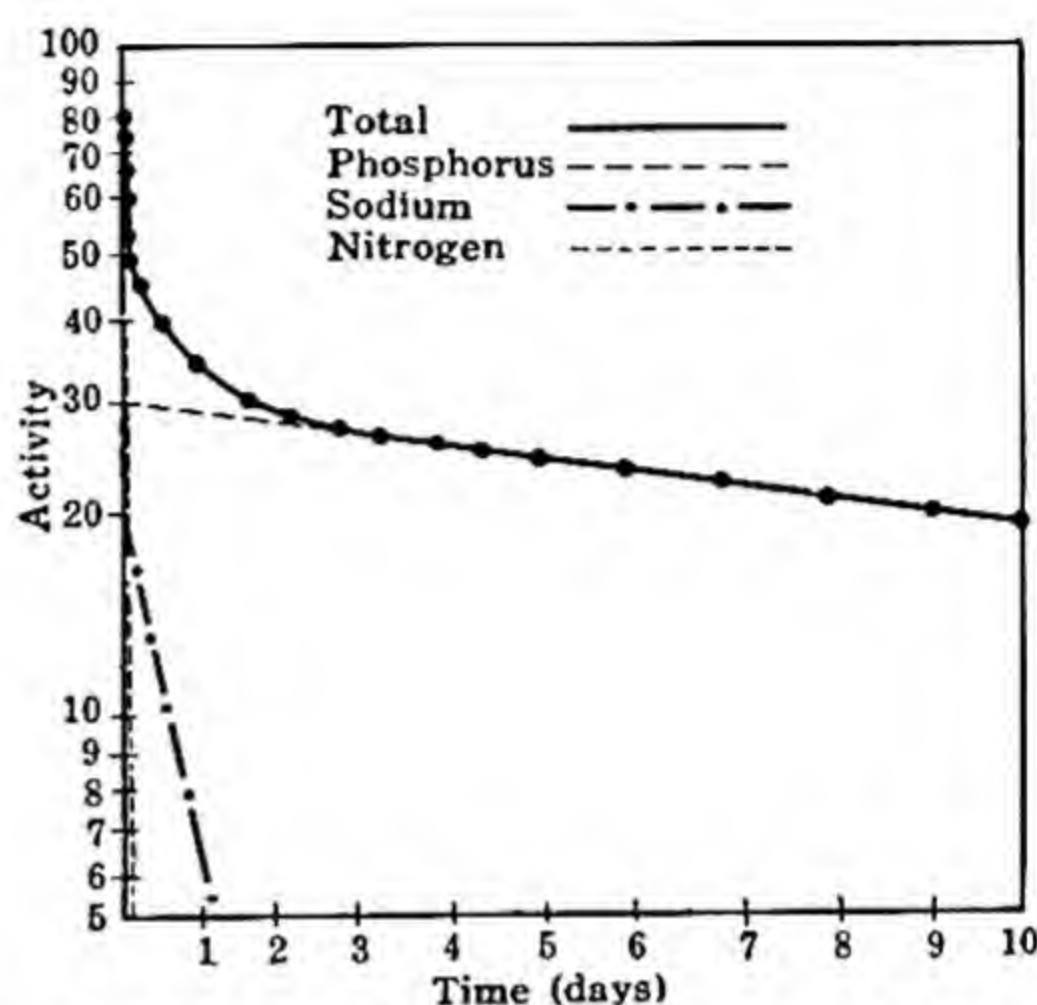


FIG. 1. Decay curve for several elements present at once, plotted semi-logarithmically. After a long time all the activity is due to the long-lived element, and by drawing the straight line back the part of the activity at short times due to this element can be deducted. This gives a second activity curve, from which the activity due to the element of intermediate half-life can be deducted. The result is three separate straight lines corresponding to the three elements present. The background activity must first be deducted as it does not obey a logarithmic decay law.



constants. It is of interest that this procedure could be applied as a method of analysis for small traces of elements. The curves of Fig. 1 correspond to an actual sample; notice that only about half the initial activity is that of the required phosphorus.

The same type of analysis can be applied to absorption curves for gamma rays; the logarithm of the activity is then plotted against the thickness of the absorber.

It may happen that the various periods are rather close in value, in which event the peeling-off process is less accurate. (It is at its best when the half-lives are quite different.) There one is up against the sort of problem faced by an analytical chemist and must seek what aids one can. A very useful technique is to follow the decay of several sections of the sample, using a magnetic field to bend out positrons for one, or a layer of lead for the other, in the hope that the various elements will have different properties and so will be detected alone by one of the detecting apparatus. Each target prepared should be analyzed by plotting a decay curve with an electroscope and a rough absorption curve in aluminum before it is decided that the sample is satisfactory for the intended purpose.

It will be realized from what follows that the manner of preparation may greatly influence the nature of the product formed, and it must be repeated that each mode of bombarding should be tested as described to make sure that the later experiments are not nullified by the presence of an impurity. To make this point clear consider numerical work with the sample of phosphorus of Fig. 1. If it were measured quickly in the cyclotron laboratory and handed over as having the full value at short times, then put into an animal and various sections of the animal brought back to the same laboratory at the end of a week, it would have appeared that much of the phosphorus had disappeared somewhere, and startling theories would be advanced to account for the loss. By taking both decay and absorption curves and looking up the values obtained in reference tables, it is easy to avoid such errors. The use of the decay curve has been illustrated above; it might be worth pointing to an example of the use of absorption. Thus, if a source of  $P^{32}$  is obtained by bombarding  $KH_2PO_4$  in a nuclear reactor, there will be present initially considerable activity from  $K^{42}$ . This gives energetic beta rays of maximum energy of 3.6 Mev, while  $P^{32}$  emits electrons of maximum energy 1.65 Mev. The presence of  $K^{42}$  can therefore readily be detected with aluminum absorbers thick enough to exclude the phosphorus radiation.



## Growth of sources under bombardment ✓

Most of the readers who have assimilated the material on the decay of sources will be able to foresee the nature of the yield obtained for various times of bombardment. There will be a steady increase in the yield of atoms available to decay until the rate of production is equal to the rate of decay, after which there is no change. The exact nature of this increase in the yield can easily be seen. We remember that there are two rates of change of the number of radioactive atoms present, one an increase at a rate depending on the beam current, and the other the decrease due to decay, a decrease proportional to the number of radioactive atoms present. With the simplifying assumption that the beam current is steady, we can represent the sum of these two rates of change as follows:

$$\frac{dN}{dt} = k - \lambda N$$

where  $N$  is the number of radioactive atoms present,  $k$  represents the constant rate of production of radioactive atoms due to the beam, and  $\lambda$  is the familiar decay constant. This equation leads to the conclusion that

$$N = \frac{k}{\lambda} (1 - e^{-\lambda t})$$

The reader will see the similarity to the growth of the current in a circuit containing inductance and resistance when the source of electromotive force is applied. It will be noticed that it requires infinite time for the final value of  $N$  to be reached but that there is 50 per cent completion after a bombardment time equal to the half-life. If we put  $N_{\infty}$  for the total possible number of radioactive atoms obtainable with a given beam, then  $N_{\infty} = k/\lambda$ , and the equation above becomes

$$N = N_{\infty}(1 - e^{-\lambda t})$$

and substitution of the value of  $0.69/\lambda$  for  $t$  as explained in the previous chapter gives the figure of 50 per cent  $N_{\infty}$  mentioned above. If bombardment for two half-lives is carried out, the percentage of the greatest possible yield is 75.

## The curie

Once the numbers formed are known, these, together with the decay constants, tell us the amount of initial activity by the simple relation



$dN/dt = -\lambda N$ . It is common to measure this activity in terms of the curie, and a discussion of this method of measurement is advisable here. The *curie*, named in honor of the discoverers of radium, refers to the amount of radon in equilibrium with 1 gram of radium element; a *millicurie* is one-thousandth of this amount. The convenient fact that radium can be detected without taking it out of its container led to the practice of comparing amounts of radium by their effect on an electroscope which was so shielded as to detect only the gamma radiation; provided that the comparison was always between radium sources the measurements had definite meaning. However, the convenience of the method of comparison has brought with it a tendency to compare sources of different elements with one another by the same means, which has reduced the term millicurie to something less precise than its true meaning. Thus it is common to speak of a millicurie of radiophosphorus, which is neither radium nor a gamma-ray emitter. If one looks at the fundamental quantity one seeks to describe about a radioactive source, it is not so much the ionization which will be produced in an electroscope, because this will depend greatly on the nature of the particular radioelement of which some eight hundred are now known, but rather the number of atoms decaying per second. This quantity can be measured accurately, and it is fair to compare two elements on this basis even if one is a beta-ray emitter and the other decays by *K*-electron capture which produces almost no ionization at all. Even if we accept this as the measure of activity it is still possible to use radium as the standard, for radium decays by alpha-particle emission and the number of atoms decaying per second can rather easily be determined by counting the number of alpha particles emitted per second by a known amount of radium, and so we can adopt a new definition of millicurie. This new definition is: *a source which decays such that  $3.7 \times 10^7$  atoms change per second is of 1-millicurie strength.*

A second unit, the *rutherford*, is now taking its chance with custom to see if it is acceptable. This is simply a name for a source which decays at the rate of  $10^6$  atoms per second. The abbreviation is *rd*.

### Example of a tracer experiment

By way of making the procedures of radioactive techniques seem more real consider an example of research which indeed needs more work. The basic problem considered is that of diffusion of various kinds of elements through cell membranes. This is an interesting and important question in biology and no adequate theory yet exists



for the process. Suppose that for cells we choose red blood cells, or erythrocytes, and we pick on sodium, potassium, phosphorus, and chlorine for study. These are available as indicated in the table.

ELEMENT	HALF-LIFE	RADIATION
Na <sup>24</sup>	14.8 hours	1.4 Mev $\beta$ ; 1.4 and 2.8 Mev $\gamma$
P <sup>32</sup>	14 days	1.65 Mev $\beta$ ; No $\gamma$
Cl <sup>38</sup>	37 minutes	4.94, 2.8, 1.2 Mev $\beta$ ; 2.15 and 1.60 Mev $\gamma$
K <sup>42</sup>	12.4 hours	3.5 and 2.1 Mev $\beta$ ; 1.51 Mev $\gamma$

Actually these are by no means the only isotopes which could be used. Na<sup>22</sup> and Cl<sup>36</sup> are available and indeed one might consider K<sup>40</sup>. These are all long-lived elements. Now it appears as though the longer the half-life the better. This is not necessarily so. In the first place the longer the half-life the less the decay constant and the more atoms have to be added to obtain a reasonable number of counts. In the second place long half-lived elements are hazards from the point of view of contamination and disposal and so are not always preferable. We will then assume that the four listed isotopes have been chosen.

The radioactive element, say Na<sup>24</sup>, is then brought into the appropriate chemical form, for example, 0.01th *M* NaCl, and appropriate strength. This is done by adding the requisite amount of inert NaCl to a solution of Na<sup>24</sup> which has the right number of counts per milliliter.

A sample of this original solution, which we call the standard, is then taken, for example, 0.1 milliliter can be drawn off, placed in a watch glass, and allowed to evaporate. The size of drop should not exceed half an inch in diameter. This is placed under a beta-ray counter of the type indicated on p. 35 or under a Lauritsen electroscope, and the activity is determined. This will be some guide to the proper conditions for subsequent counting. Usually it is wise to put the sample at least 1 inch from the counter opening.

The radioactive solution and the erythrocytes in whatever biological form is under study are then mixed for a definite time and the erythrocytes removed by centrifugation, washed thoroughly, and placed in a watch glass which is put under the counter. The total weight of red cells should be taken. If this process has taken more than an hour the time should be noted at every stage so as to allow for a correction for decay.

The count is then recorded and this, less the background count, is the activity. This can be compared with the original activity and



the amount diffused or absorbed can be at once seen in relative terms. Repeating the process for different times of diffusion enables a permeability constant to be measured.

The same procedure can be followed for potassium and phosphorus. For chlorine a decay correction is necessary. This is done as follows. The time of every count is recorded and a table made as below.

TIME OF DAY	ELAPSED TIME (min)	FACTOR	COUNT	TIME (min)	NUMBER PER MINUTE	NET PER MINUTE	CORRECTED
11:02		1.00	532	1	532	522	522
11:37	35	0.52	75	1	75	65	125
11:43	41	0.46	88	2	44	34	74
12:01	59	0.33	98	2	59	49	148
12:09	67	0.285	161	1	161	151	530
Background, 10 per minute							

The factor to allow for decay is determined by plotting on semi-logarithmic paper a straight line passing through the 50 per cent figure at the correct half-life and reading off the amount by which the source has decayed at the time of the count. By dividing the actual count by this figure the activity at the time of no decay can be found. This gives the corrected figure which can then be readily compared.

A simple device which the authors have used is to put a logarithmically graduated dial appropriate to the element on an electric clock and to read the factor directly.

It is wise to check periodically the original sample made from the first solution. This is a safeguard against variation of the counter and circuit. Also the background count should be taken from time to time.

If it is necessary, after the counting process is completed, to know the actual amount of material which has diffused, it is best to know the total amount of an element, inert plus radioactive, in the original solution. Then the amount in the standard can be computed purely in terms of the volume of the original solution and the volume sampled. It is possible to figure the number of radioactive atoms from the counting rate and the solid angle and efficiency of the counter, but this is not usually done. The amount of inert contaminant generally far exceeds the amount of purely radioactive material and hence the calculation is meaningless. However, it can be assumed that in the vast majority of cases the radioactive and inert material in the original solution will behave in precisely the same way so that the radioactive count can be taken as a measure of this mixture.



In this way the measurement of very small rates of diffusion can be made.

This short account has not considered the permissible error limits of the experiment. Taking the square root of the count in the table gives an estimate of the order of fluctuation to be expected. Thus with the counts as we have indicated an overall accuracy of 10 per cent is all that can be expected.

### Preparation of radioelements

The great majority of radioelements in use today are made in a chain-reacting pile, or nuclear reactor. The reason for this is the fact that neutron reactions have high cross section and that large amounts of neutrons are available in a pile. The considerations in the manufacture of radioelements are not really within the scope of this book as they present a problem related in detail to the working of the pile. The kind of factors to consider are, for example, the way in which the introduction of non-fissionable material affects the pile operation. Clearly, too much will stop the chain reaction from progressing. The form in which the substance is irradiated is important; it must not react with the container either before or after transmutation. It is also necessary to avoid excessively strong sources as they form a radiation hazard and cannot cheaply be sent by express.

An excellent working system for pile irradiation and distribution has been set up by the Atomic Energy Commission in the United States, and a similar system operates at Harwell in England. A catalogue of available isotopes can be had on request from the Atomic Energy Commission, Isotopes Division, at Oak Ridge, Tennessee.

The kind of nuclear reaction used in the pile is important. The cheapest is the fission reaction itself. This is always happening, and the radioelement is thus a by-product of pile operation. This reaction is responsible for the formation of  $I^{131}$  for example, and since iodine is fairly easily chemically separated it is readily available and cheap.

Slow-neutron capture reactions are next in availability. Some of these have very large cross sections and yield large amounts of radioisotope. The product is almost always the same as the element bombarded which brings up a very important secondary point, that of *specific activity*.

It is very inefficient to try to convert the whole of a target into a radioelement for very often the resulting activity is far too "hot" to handle and also the number of inert atoms under bombardment gets less as time goes on. So if it is required to have a radioelement



which contains very little inert element, and this is very often essential as too great a mass of an element may change the metabolism of a plant or animal, it is necessary to find some way around the limitation mentioned. One very good way is to use a reaction which starts from a different element. For example,  $P^{32}$  can be made either from ordinary phosphorus,  $P^{31}$ , or from  $S^{32}$ . The former yields a low specific activity. The latter, since phosphorus can readily be chemically separated from sulphur, yields high specific activity. The reaction used is an  $(np)$  type,  $S^{32}(np)P^{32}$ , and requires relatively fast neutrons. Since pile neutrons are predominantly slow the reaction efficiency is less and  $P^{32}$  of such high specific activity is more expensive. In some cases  $(n\alpha)$  reactions can be used, such as  $Ca^{40}(n\alpha)A^{37}$ .

The neutron flux in a nuclear reactor runs around  $10^{10}$  per square centimeter per second, which is the figure quoted for the small reactor at Harwell. This gives some idea of the possible rate of formation of a radioactive element. Taken in conjunction with neutron cross section figures a tolerable estimate of the yield can be made.

A certain amount of cyclotron bombardment is still called for. Elements which are made by decreasing the neutron content, which includes the large class of positron emitters, are made by cyclotron bombardment. Thus  $Na^{22}$ , a 3-year half-life positron emitter, is best made by the  $Mg^{24}(d\alpha)Na^{22}$  reaction, using a cyclotron having high-energy deuterons.

Cyclotron-produced radioelements of tolerably high specific activity can be made. This is because the penetrating power of deuterons is not great and a target, say  $\frac{1}{20}$  cm thick, is wholly adequate. A rather high activity can often be built up in such a target even though chemical separation is not used. The highest specific activity still requires chemical separation.

Cyclotron bombardment involves a considerable cooling problem because the target is normally bombarded in a vacuum and the heating is confined to a very small surface layer. Moreover if the target volatilizes at all it may ruin the cyclotron vacuum and also contaminate the inside of the vacuum chamber. To overcome these difficulties the target is made thin and inclined to the beam so as to offer the maximum area for cooling.

Designing a target is a good exercise in physics. Where good conduction is possible quite strange targets are practicable. Thus metallic sodium can readily be bombarded if the backing metal is water cooled, whereas sodium fluoride, which has a high melting point but conducts poorly, will melt and vaporize.



If the beam is brought out through a thin foil window, bombardment is of course much easier. Usually much beam is lost in the deflection system, so large sources are more expensive to make in this way. A good-sized cyclotron costs \$10 per hour to run, so sources are never really inexpensive.

Generally speaking, any metal can be bombarded in a cyclotron. Iron phosphide can be used as a target for iron and phosphorus but is hard to get in solution. The right form can be dissolved in aqua regia and the chemical separation is straightforward. Other targets are specialized and need expert advice.

A word about the bombardment of compounds is in order. It may seem that the bombardment of, say, tobacco mosaic virus by neutrons might give viable and radioactive virus. Whether this is possible with a low yield is of great interest, but it can be seen clearly why the yield will not be high. The capture of a neutron gives a nucleus about 8 Mev of excitation energy. Now even if the bombarding neutron is slow, this 8 Mev must be lost as gamma radiation before the final radioactive nucleus is formed. The process of emission of the gamma ray causes the nucleus to recoil, and this energy of recoil amounts to about 100 electron volts. The energy binding two atoms in a molecule seldom exceeds 5 electron volts so that the process of recoil has much more energy than the chemical bond and the result is therefore a disruption of the molecule. The chance of a return to the original configuration is rather low so that the result of such a bombardment is to create radioactivity but not the proper molecule.

In some cases the yield of properly "tagged" molecules may be higher than expected; for example, radioactive cystine has been made in this way by bombarding cystine with slow neutrons. The explanation is probably that the 8 Mev of excitation energy can be lost as two or more gamma rays in cascade. For two equally energetic gamma rays there need be no nuclear recoil at all. Hence for strong chemical bonds and nuclei rich in energy levels there is a decided possibility of making radioactive elements by this means.

It can be mentioned that the fact that the recoil dislodges the newly radioactive atom can be used to concentrate radioactive sources. The method, devised by Szilard and Chalmers, is to irradiate an organic compound, say ethyl bromide, and create considerable radioactive bromine which is freed from combination. Normally this would recombine but by supplying an excess of free bromine the recombination favors the inert substance and the radioactivity concentrates in the free bromine. The amount of free bromine added, though large



compared to the radioactive bromine, is still small compared to the bound bromine in the ethyl bromide. This is why the method works. The Szilard-Chalmers process has lost much of its importance since extensive neutron irradiation is now easy. It played an important part when the relatively small neutron flux from a cyclotron had to be used.

The commonly used radioactive elements and their preparation are listed in Appendix 3. For details of the preparation of any source the reader is referred to original literature.

### Precautions in using radioactive substances

The first and most vital precaution regarding radioactivity is to avoid getting any of it inside the body. A microcurie of long-lived radioactivity inside is as dangerous as a curie outside if allowance is made for the fact that the curie outside is handled only for part of the time whereas radioactivity in the bones is at work day and night. Not all elements are equally dangerous, and short half-lives are of course much safer. Nevertheless, it is well to treat radioactivity as a bacteriologist treats bacteria, to learn to keep one's fingers away from one's face, and to avoid smoking while manipulating radioactive materials.

In the present state of the art it is not very informative to try to quote tolerances for various radioelements in the body. By way of an example the figures computed by Morgan for  $I^{131}$  and  $Ca^{45}$  are given. For iodine which concentrates 20 per cent in the thyroid, 2 microcuries will give an initial tolerance dose. For calcium which concentrates in the bones, a much more distributed locale, the same dose requires 190 microcuries. To produce an average tolerance in a year, however, 960 microcuries are needed for  $I^{131}$  and only 400 for  $Ca^{45}$ , the reason being the longer half-life of the calcium.

It is apparent that quite small amounts are adding significantly to the radiation hazard of the individual.

The second precaution is to avoid external radiation of too great intensity for safety. This is discussed more fully in the next chapter, but it can be pointed out that a millicurie of any source which emits gamma rays produces one-tenth tolerance radiation at 1 meter. This is a very rough figure but is useful for estimate. An uncovered beta ray emitter is about 200 times more dangerous. Therefore beta-ray sources must always be handled with tongs for very short periods and must be put behind shields as rapidly as possible. Burns on the hands from beta rays are all too easy to produce.



Actually the great majority of actual tracer manipulation is with sources of less than a hundredth of a millicurie. Not much danger is encountered from these, particularly if one follows the practice of leaving the counter or electroscope to record and going away while the actual count is being taken.

The third precaution is to avoid spilling and contamination. A place must be made for old radioactivity, either a special sink for short half-lived material or a cavern for long half-lived waste material; and rigid discipline must be observed to see that all dead experiments are consigned to one of these places. Spilling always occurs, so provision, preferably in the form of disposable table tops of thick card or linoleum or thick paper, must be made for cleaning up after a spill. Any disposal unit for radioactive sources must be clearly marked. Rubber gloves must be worn for all manipulations, and it is wise to use a hood for every doubtful case where powders can be blown around. Hands must be thoroughly washed in the special sink.

Contamination is not so much a health hazard as a laboratory nuisance. Many experiments cannot be made at all with a high background count. Therefore the existence of contamination should be in a class with the existence of dandelions in the lawn and treated with the same ceaseless vigilance.

If sources which definitely present a radiation hazard are to be used, then the entire plan of procedure throughout the experiment must be thought through so that no operations are made on the spur of the moment. Usually the important considerations are storage and manipulation in the initial stages before the source has become dispersed in solution or through an experimental system. Storage involves the problem of shielding (which is easy to calculate) and the more difficult question of handling while taking the substance in and out of storage. Varying degrees of remote-control equipment are needed for this. To some degree existing chemical tongs and beaker-tongs designed for thermally hot objects are suitable for radioactively "hot" objects so that equipment to handle up to 50 millicuries for a few seconds can usually be bought and improvised easily. One or two lead shields an inch thick and several inches across can be used as a barrier to protect the hands and nearby body regions during such manipulation. Processes such as pipetting, mixing, and filtration each require remote operation if the sources are from 1 to 50 mc, and existing laboratory equipment usually can be adapted to this.



The most important precaution is to use some monitoring equipment *which is capable of detecting the radiation* to check every operation which can conceivably be dangerous. Such equipment is described in the next chapter.

### Summary

To summarize this chapter: We have considered the types of nuclear radiation and the detection problems arising, pointing out that beta rays are absorbed by the rough rule  $t \times d = 0.54E_m - 0.16$ , and giving an account of the absorption of gamma radiation; we have also appraised counters and electrometers, indicating their strong and weak points. The problem of dealing with a mixture of decay periods (or absorption coefficients) has been considered and the method of procedure outlined, after which the way in which a source grows under bombardment was explained and actual examples considered. A millicurie was defined as a source which decayed at the rate of  $3.7 \times 10^7$  atoms per second, and the methods of preparation of the more common radioelements were outlined. A short account of a possible tracer experiment has been given with some brief advice on how to handle radioactive material both for health protection and for the reduction of contamination.

The chapter might, in mathematical language, be described as "necessary but not sufficient." There has been a sense of completeness in the authors' minds as the previous chapters were written, but here that sense is lacking. It is certain that many new methods of concentration will be developed, that many more radioactive elements will find application, and that better bombardment techniques will soon be forthcoming. However, this book does not aim at being a complete reference source; it is intended to make available many known facts in nuclear physics to the people who are advancing the subject of applied nuclear physics, and to make others appreciate their work. This little postscript leads very nicely into the following chapter, where we move right up to the frontier; and while we attempt to describe some of the experiences on that frontier we have some of the troubles of frontier days in that all the news of discovery does not get away from its locale fast enough and the account we give of actual experiments will be rather like the tales that filtered back to the East from travelers in the West a hundred years ago—interesting, but not a complete story. It should be read in that spirit.



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## 8. Artificial Radioactivity in Practice

*A space whose every cubit  
Seems to cry out . . .*

The study of the nucleus of the atom is one of the newest and most rapidly advancing in the whole subject of modern physics. Yet in writing thus far we have had a feeling of relative stability; a feeling that we have not omitted a great deal of the content that a reasonably satisfactory account of the nucleus should have. This feeling is totally lacking as we begin this chapter. Instead we have the feeling that we are here starting an account which cannot but be incomplete, that we are putting a heading on a single chapter which requires several volumes to describe. This being so, we wish to make it clear that this chapter is intended only as a collection of illustrative experiments which may make it easier for some specialist in a field of work to devise his own experiments and to add his quota to the volumes of important literature to be accumulated in the next decade.

We can make a rough division of this chapter into tracer work and other, and begin by considering the problems involved in the former.

### Tracer experiments in general

There is only one feature common to all types of tracer experiments and that is the obvious one of preparing the radioactive form of the element to be used. This is the problem that has to be solved by the nuclear physicist, and we can here suppose that it has been solved. This means that either a container arrives from pile irradiation with the expected material, or a cyclotron-bombarded target is made available. The purity of both must be considered, more particularly of the latter because sources from cyclotrons are not usually made under such controlled conditions as in a pile.

His next problem is to get the radioactive material in the right form for introducing it into the system he wishes to study. This stage is one of the severest "bottlenecks" in the progress of tracer experiments



and should be considered most seriously immediately after the idea of using radioactive materials has occurred to the experimenter. It may happen to be relatively easy; for example, if one wishes to use sodium or potassium in water solution, all one has to do is to assure oneself that the physicist in charge of the cyclotron has not included a large amount of copper along with the sodium, for example, and then dissolve the sodium salt in water and go ahead. On the other hand the biochemist may have great curiosity about the way in which a hormone produces growth: where does the original carbon of the hormone go after the growth has progressed? The answer to this question could be obtained by using radioactive carbon, but it would mean the synthesis of an extremely complicated organic molecule from carbon dioxide.

Rapid advance has already been made in the preparation of labeled compounds. A list of available organic compounds suitably labeled is issued by the Atomic Energy Commission and is steadily growing. The cost of such materials is naturally high, but often unique and penetrating researches are made possible by their means. The list includes glycine, alanine, calcium glycolate, and radioactive algae.

A method having been devised for introducing the radioactive element in the correct form into the experimental system, the next question that arises is, what is the dilution that will occur? Thus if a straightforward chemical extraction is to be carried out the amount of the original material in the final volume which is placed near the detection apparatus will generally be somewhere around a tenth of the original material, whereas if a cocktail of some radioactive material is given to a human being the amount detectable is likely to be in the proportion of the volume of the cocktail to that of the individual, and that is of the order of 1 part in 10,000. These figures are intended only to illustrate; they emphatically do not give even a rough guide, for the amount of material at the end depends markedly on the chemical and physiological behavior of the element under the conditions of working. Often the dilution factor can be determined only after trial and error, but it is clearly wise to start biological work with mice rather than cattle. The need of a large dilution factor should not discourage the attempting of an experiment. A very ordinary source gives  $10^9$  counts per minute, and it is not so difficult to determine 10 counts per minute with a Geiger counter, so that a dilution factor of  $10^8$  can be handled.

With these preliminaries the actual experiment can be carried out and the events studied by counting the pulses from the tagged element. Any carefully chosen method of operation which takes account of the



decay of the source, the variation of sensitivity of detection, and the reduction of counts due to absorption should be satisfactory. It is wise to duplicate all the conditions in observing the radioactivity, so that counting at the start and at the end will make a true comparison. Thus, if work were being done with  $C^{14}$ , great care would have to be taken to ensure that in each sample counted the same percentage of the beta rays, which are so easily absorbed, were being detected. It is an excellent idea to spend some little time carrying out trivial experiments, like simple dilution, to make sure that consistent numerical results are obtained with the equipment used for counting, before the actual experiment is made. On the surface, radioactivity offers speedy results—in fact, that is one of its main attractions; but, unless the experimenter takes care to become familiar with the features of the new technique of observation, it has its pitfalls.

We can mention here some of the experimental conditions to watch. First, the position of the test substance with respect to the detection instrument must be the same. Second, all samples must be prepared in the same way and must have the same thickness and material composition. If this is not possible, careful calibration with known amounts of material must first be undertaken. The consistent operation of the counter or electroscope must be tested during the experiment from time to time with a standard radioactive substance such as uranium glass or a clock face. In this connection it must be remembered that no electroscope has a truly linear scale, and that a Geiger counter may not be consistent at high rates of counting, particularly just before the rate at which the mechanical counter “blocks off.” All these difficulties can be overcome by sufficiently familiarizing oneself with the counting apparatus before use.

## ILLUSTRATIVE EXPERIMENTS

### Turnover studies

One very important series of experiments concerns the rate of turnover of various radioactive elements in living systems. This turnover has proved to be exceedingly rapid in many cases, even where the form in which the radioactive substance is examined is relatively complicated. The underlying reason is that the process of life is one of continual change, of build-up and consumption, and not in any part an inert process biding its time. This very remarkable property of life is shown with great clarity by radioactive experiments. For ex-



ample, Hevesy found that, four hours after a meal containing  $P^{32}$ , 48 per cent of the  $P^{32}$  recovered was in the bones, 25 per cent in the muscles, 2.4 per cent in the blood, and less than 0.1 per cent in the brain. The rapidity with which  $P^{32}$  is found in the bones is striking. Of interest is the uptake of phosphorus from radioactive phosphate by bacteria. This is quite rapid with living bacteria, but dead bacteria were found by Goldwasser to take up very small amounts of  $P^{32}$ . The

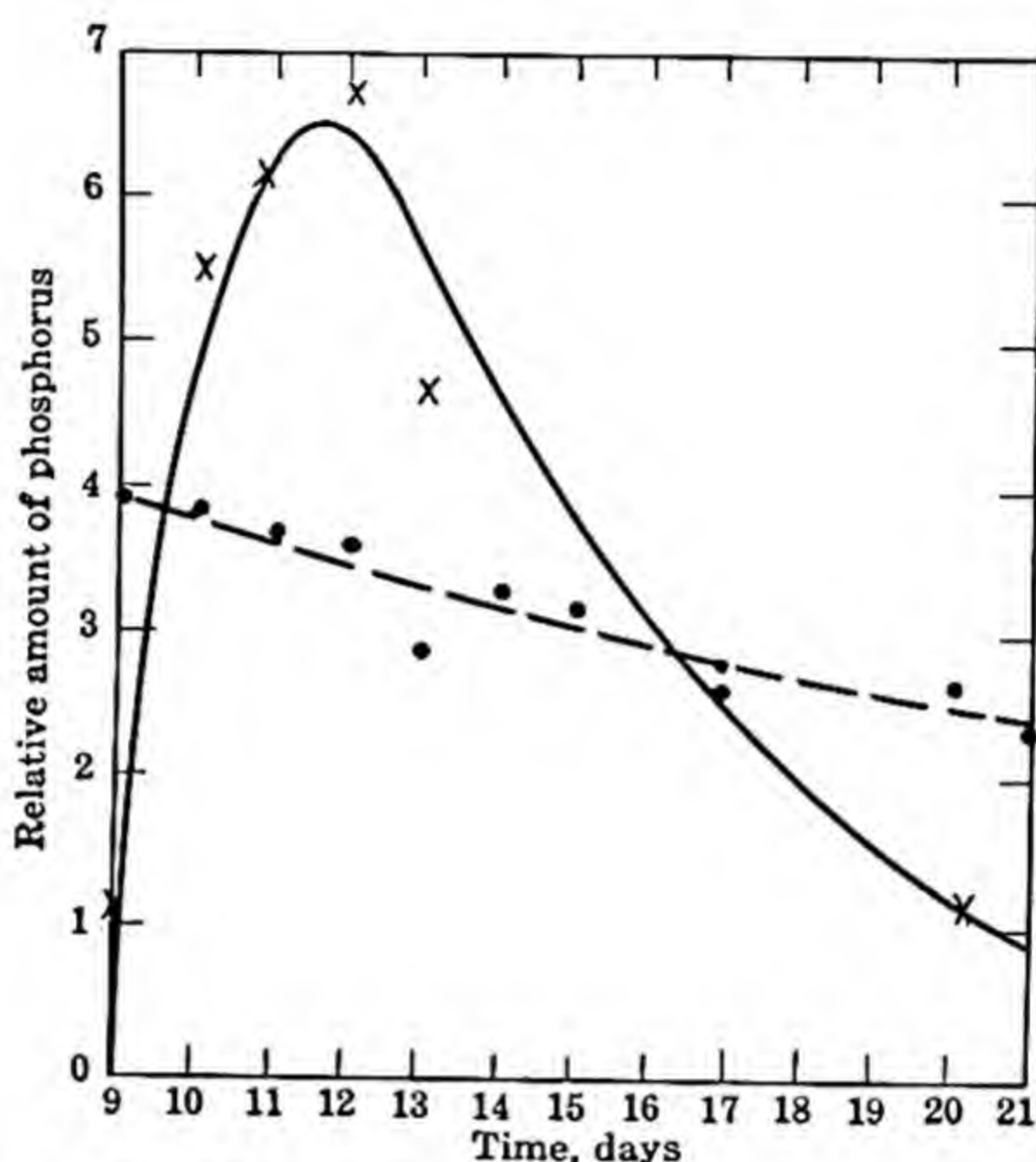


FIG. 1. Turnover curve for  $P^{32}$  in chick embryo liver as measured by Shrigley. The dotted curve gives the amount of total phosphorus, which clearly behaves quite differently from the inorganic phosphate containing the  $P^{32}$ . The curve drawn through the  $P^{32}$  points is the curve  $y = A(e^{-0.28t} - e^{-0.56t})$  so that the uptake coefficient is 0.28 per day and the turnover coefficient is 0.56 per day.

contrast between inert diffusion and living metabolism therefore involves a factor of fifty at the least.

The normal process of turnover involves a curve as indicated in Fig. 1. The rising part of the curve corresponds to the formation of a precursor from which the substance investigated is formed. Thus in Fig. 1 the points on the curve represent the amount of  $P^{32}$  in the liver of a chick embryo, the data being due to Shrigley. The precursor of phosphorus in the liver is presumably phosphate in the blood, and the rising part of the curve corresponds to the transfer of this phosphate to the liver by the blood and the conversion of phosphate, for example, to phospholipid by the liver. The falling part corresponds to the transfer of phosphorus out of the liver. Such curves can be analyzed



into two exponentials, with factors in the exponential terms which express the two processes occurring. The relation which holds is

$$y = A(e^{-\lambda_1 t} - e^{-\lambda_2 t})$$

$y$  is the amount of radioactivity measured after time  $t$ .  $A$  is a constant determined by the size of the organ, countersensitivity, etc., and  $\lambda_1$  and  $\lambda_2$  are constants of metabolism.  $\lambda_1$  is an uptake coefficient, probably related to rate of blood flow;  $\lambda_2$  is a turnover coefficient, related to the rate of transfer of the radioactive element.

The study of factors which alter the turnover figures is of great interest. Thus Stillman has shown that the adrenal gland has no effect on the formation of phospholipid in the liver; on the other hand Perlman and Chaikoff present evidence that choline speeds up the process.

Turnover studies of iodine in the thyroid are most illuminating. The concentration of radioactive iodine by this organ is indeed impressive. Hertz, Roberts, and Evans found that the thyroid concentrates 80 times as much per gram as the remainder of the body tissues. The iodine appears initially as iodide, and the quantity  $\lambda_1$  for iodide is very large. However, if the turnover of thyroxine is measured  $\lambda_1$  is found to be very much smaller, indicating that one or more precursors have to be formed before thyroxine is produced. The rapid uptake of iodine does not therefore mean that the physiological function of iodine is rapidly fulfilled, nor is it a guarantee that it will be fulfilled at all unless the process for the formation of thyroxine is functioning smoothly.

An interesting, nearly closed, biological system is an incubating egg. Studies on phosphorus turnover in eggs have been made by Shrigley, a sample result being shown in Fig. 1 which summarizes preliminary studies on turnover in the liver. The radiophosphorus was injected as phosphoric acid into the yolk, and studies of the distribution of total phosphorus and radioactive phosphorus were made at different stages of incubation. It can be seen that the phosphoric acid injected follows a different pattern from the total phosphorus, and in particular nearly all this phosphorus has been taken up by the embryo a week before the completion of incubation.

### Radioautographs

A powerful method of studying the localization of radioactive material is the radioautograph first employed by Lacassagne and Lattes in 1924. The procedure used is to give rather considerable doses of



radioactive elements and to cut sections of the organ or substance under investigation after a suitable time has elapsed. These are then placed on x-ray film with a thin film of mica or aluminum between (this is not needed if the section is perfectly dry) and exposure allowed to take place. A detectable effect is produced by  $10^6$  fast electrons and good blackening by  $10^7$ . The new Eastman Kodak emulsions with sensitivity to single electrons may find application in this work: the tracks need microscopic examination for their detection.

On development, the localization of radioactivity can be seen. Examples of this rather striking technique are shown in Plates II, III, and IV. The method is limited by the fact that beta rays are emitted in all directions and no feasibility of focus appears to exist except in one or two cases, because the beta-ray spectrum comprises many energies. Where a strong line due to internal conversion exists (as for iodine) there is a possibility that an enlarged image can be obtained. So far this has not been done. By using very thin emulsion reasonably good location of the radioactivity can be achieved.

This method has produced some striking results. The concentration of radiostrontium in developing bone (including bone tumors) has been shown by Pechar. The histology of the thyroid has been considerably advanced by Gorbman, using radioautographs of iodine. Bowen has studied the barium cycle in hornets by this method. It can be applied with great success to locate the various components in paper chromatography.

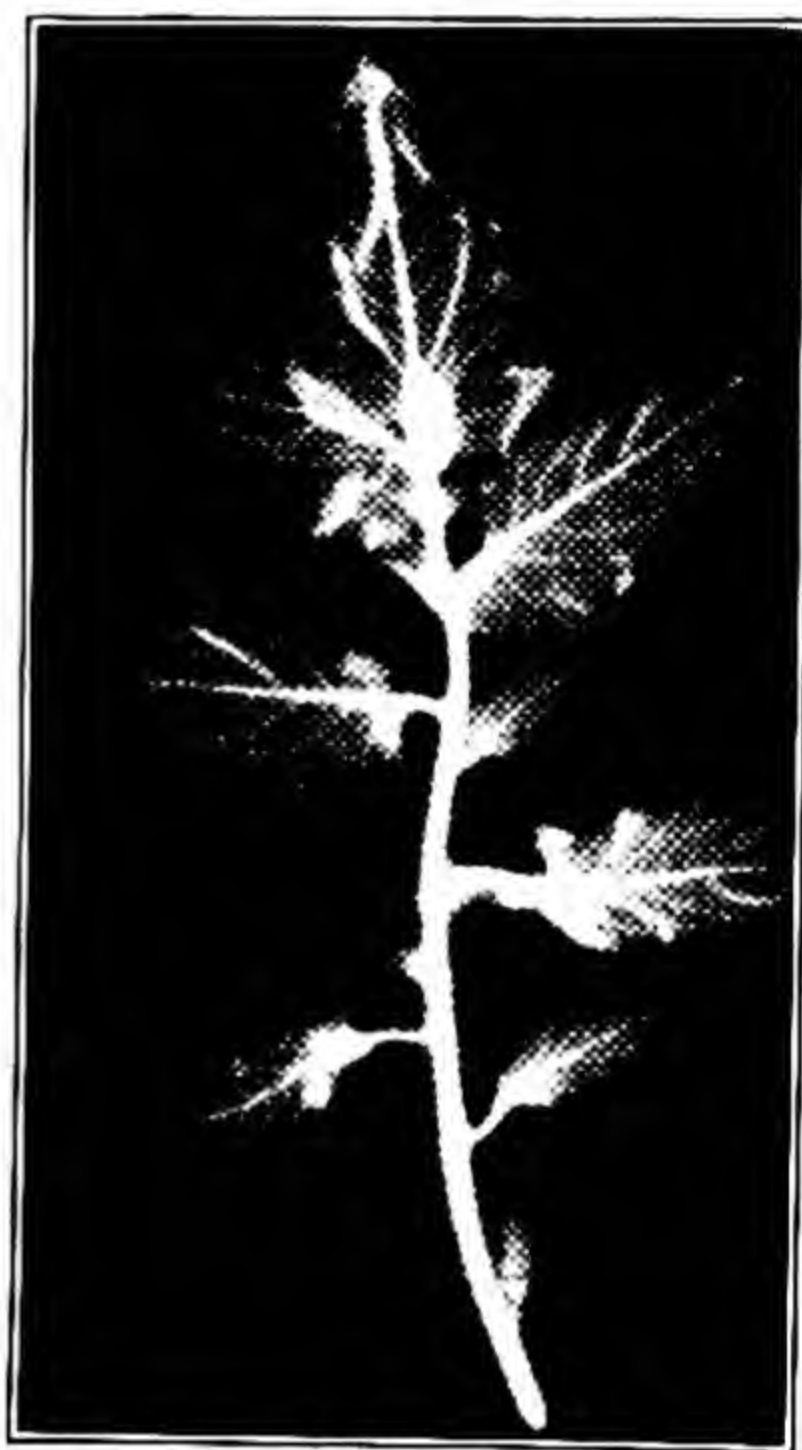


PLATE II. Radioautograph, taken by Dr. J. G. Hamilton, of the distribution of radiophosphorus in the leaves of the tomato plant. The accumulation of phosphorus is shown by the light areas.

### Diffusion and exchange studies

A very straightforward application of radioactivity is in diffusion studies. Somewhat similar experimental work can be done on exchange processes, which are indeed rather like diffusion.

Gaseous diffusion involves a rather complicated theory. This theory is much simpler when the diffusing gas molecule is the same as the



gas through which it diffuses. Until the advent of radioactivity or of the detection of isotopes by the mass spectrograph, no method was known to test this theory. Using these techniques several tests have been made, for example, the observation of the diffusion of  $A^{41}$  into argon by Hutchinson. The results are interesting in that the diffusion constant at a variety of temperatures was measured and evidence regarding the force between two argon atoms obtained.

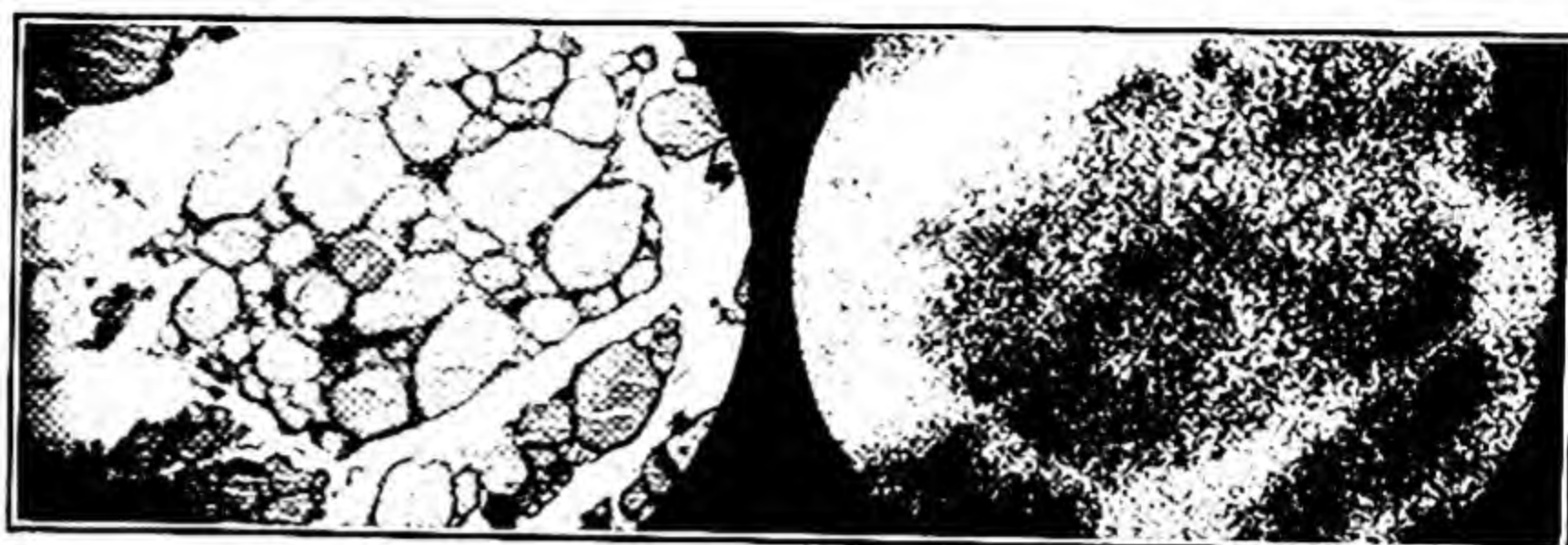
Whether diffusion occurs at all from the lungs to the blood has been tested by using radioactive krypton and argon. In spite of high concentrations of these gases in the lungs no radioactivity was observed in the blood by Jones and a group of workers who were studying the process of gas bubble formation in the blood. The uptake of oxygen and nitrogen by the blood is therefore chemical and not physical.

"Diffusion" into red blood cells has been investigated by Smith, Winkler, Eisenman, and Ott, who tested the amount of radioactivity held by erythrocytes after exposure to radioactive sodium, chlorine, phosphorus, and potassium. The uptake of phosphorus was clearly not a diffusion process since at low temperatures the "diffusion" practically ceased. This could not be said so definitely for the other three elements. Brooks has made further studies on this.

Exchange is a problem of central importance in radioactive tracer work. It can be seen most clearly where ionization takes place, for example, for NaCl. For this particular compound, which is formed because of the attraction between  $Na^+$  and  $Cl^-$ , the action of the high dielectric constant of water is to cause separation of the two ions, which are then free and stable because of the grouping of polar molecules of water around them. Such ions are individuals and free to wander and to reassociate anywhere they please. Clearly there can be no permanence whatever about any molecule of NaCl once it is in solution. This extreme case of *exchange* has a counterpart in other molecules, even though the process of ionization does not take place. A moment's thought will show that the bombardment of a molecule by one of its constituents is rather different in character from a bombardment by, say, water, or by some completely different element. Thus, consider  $CH_3I$ , which has been studied by McKay. If the methyl iodide is bombarded by iodine molecules there will be a disruption of the molecule at the time of the collision, and this will be followed by a return to the stable configuration of  $CH_3I$ . However, the whole requirement is satisfied by the formation of  $CH_3I$ , and it makes no difference whether the original iodine atom returns or a different atom. Chemically there is no way to tell that any change has



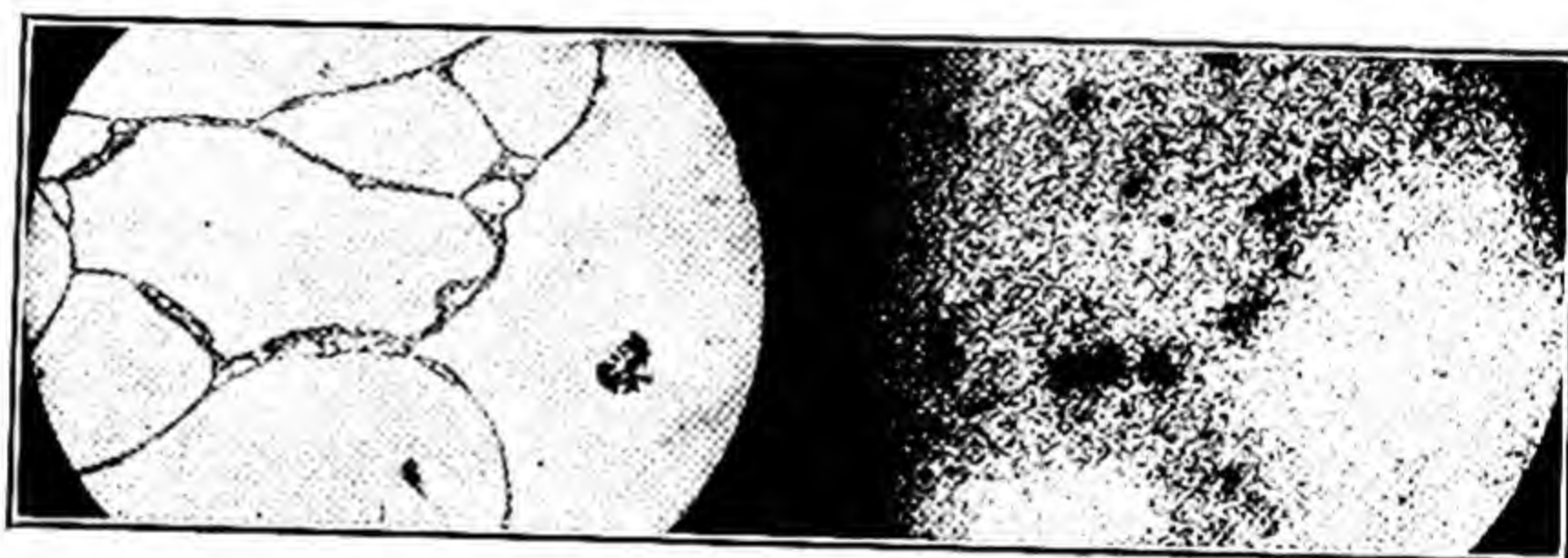
Normal thyroid tissue. Photomicrograph on left, radioautograph on right. The darkened areas in the radioautograph represent the regions of greatest accumulation of radioiodine.



Hyperplastic thyroid. The section on the left shows large grayish areas which contain colloid, while the remaining acini are devoid of colloid. The radioautograph indicates that most of the accumulated iodine was stored in the colloid.



Non-toxic goiter. Photomicrograph on left, radioautograph on right. The acini are enlarged and distended with colloid which has accumulated very little iodine. The cells and small acini surrounding the colloid deposits stored much more iodine.



Cancer of the thyroid. There are three small islands of uninvaded thyroid tissue which can be seen from the radioautograph to have accumulated most of the radioiodine.



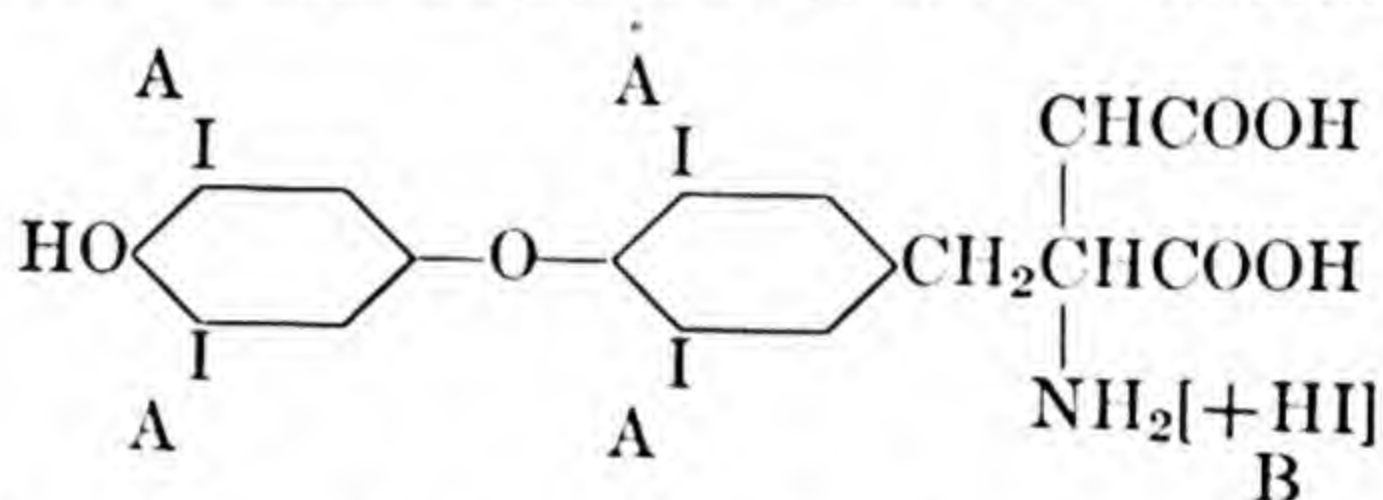
PLATE III. These photographs were made by Dr. J. G. Hamilton, and the legends are virtually those given by him in his article in the *Journal of Applied Physics*.



occurred. However, if the bombardment is by means of radioactive iodine, the radioactivity in the free iodine diminishes and that in the methyl iodide increases, owing to the exchange process.

The reader can see the two major factors which determine whether exchange will take place. The more strongly bound an atom is to its molecule, the less will it exchange. The greater the energy of molecular bombardment, the more the exchange. This last is determined by temperature, and accordingly faster exchange will be expected at higher temperatures. This is found to be the case.

In many tracer problems it is essential that exchange be impossible, or nearly so. This leads to the need for preliminary research as to whether exchange can take place. Such experiments are direct and almost physical in character in that all that is required is the test of whether molecular bombardment affects exchange. Thus, if a phospholipid is shaken with a radioactive phosphate there is found to be no exchange of the phosphorus, so that the simple bombardment process we have described does not take place. The location of a radioactive atom in a molecule is of great importance. Thus, in thyroxine, radioactive iodine attached as at A will not exchange, whereas



attached at B as the hydrogen iodide it does exchange.

One can say roughly that for hydrogen, carbon, nitrogen, phosphorus, and sulphur exchange is no great problem, whereas for the halogens and alkali metals it is serious.

### Dilution studies

A by no means trivial technique involving radioactive tracers is the measurement of total volumes or total quantities by dilution measurements. Thus, if, say,  $10^6$  counts per minute of radiosodium are injected into the blood stream of an animal and a few minutes later a known volume  $v$  of blood is removed and found to give  $x$  counts per minute, it is possible to determine the volume of blood in the animal. Let this be  $V$ ; then

$$x = \left( \frac{v}{V} \right) 10^6$$

which determines  $V$ .



Such simple measurements can be extended to the measurement of the total amount of a specific compound in the blood. Here one injects a known amount of the compound carrying some radioactive tracer element and recovers a known amount of the compound after mixing has taken place. The same equation now holds for this specific compound. It is not confined to biology, of course, but can be applied to any large and unmanageable mixture provided it is possible to secure complete mixing and to separate a known amount of the compound of interest.

### Special problems

By way of illustration we can consider the application of tracer technique to one or two detailed problems. As a first example we can consider the problem of vitamins in nutrition. An important constituent of corn is phytic acid which is a hexahydroxycyclohexane in combination with phosphoric acid. Phytic acid is an important source of phosphorus for poultry, but the bird does not make use of the available phosphorus unless at the same time it has one of the antirachitic vitamins, the vitamin D's, in its diet. Several such vitamins are known, notably those derived from ergosterol and cholesterol.

Experiments by Singsen and Matterson showed that for young turkeys the cholesterol-derived vitamin D is more effective in promoting uptake of phosphorus from phytic acid than the ergosterol variety. A test of this can be made with radioactive phytic acid, the experiment being to feed turkey poults radioactive phytic acid and various vitamins, then to take samples of blood after various intervals and look for the amount of radioactive phosphorus in each. An increase after feeding a particular vitamin is evidence that such a vitamin is more effective.

The experiment thus has two parts. The first is the production of radioactive phytic acid; the second, the assay of the radioactivity in the turkey poults. The radioactive phytic acid was produced by biosynthesis; that is to say the metabolism of a plant was relied on to put radioactive phosphorus into the form of phytic acid. To do this twelve corn plants were cultivated at the Connecticut Agriculture Experiment Station by Dr. Jacobsen until the stage of ear formation was beginning. The corn plants were grown in water and the diet was made deficient in phosphorus for a while; then 200 millicuries of potassium hydrogen phosphate were added to the water. The result was indeed sensational, for in a day or two this activity was spread all over the plants (a feature which should be remembered in taking



safety precautions, for the plant offers little absorption for the beta rays, while the water from which they are growing completely stops the radiation). A reasonable amount went into the ears and sufficient phytic acid could be extracted from them to continue with the second part of the experiment. It is likely that an increased yield would have been obtained if the plants had not been starved of phosphorus be-



PLATE IV. Radioautograph made by Dr. Perry Stout of the Department of Plant Nutrition of the University of California, showing the distribution of radioactive zinc in the fruit of a tomato plant which had been fed radioactive zinc chloride. The slices of the fruit were placed on a photographic plate, and the beta rays from the zinc caused action in regions where zinc was concentrated. It can be seen that the zinc concentrated in the seeds.

fore, because the normal metabolic process is at its maximum in the ear and this would have taken most of the new phosphorus. However, this is not certain.

The second phase was carried out by separating the available phytate into equal fractions and feeding it to selected poultts with known vitamin histories and with controlled amounts of vitamin in the diet. The result obtained is typical of tracer work. The first day's observation of the uptake of phosphorus followed the expected pattern, and indeed the cholesterol-derived vitamin produced increased radioactivity in the blood samples taken. However, it became clear as time went on that the interchange of phosphorus between inert phosphorus



in the bird and the radioactive phosphorus became the predominant factor so that studies of blood activity were no longer indicative of the uptake of radioactive phosphorus but rather of the process by which the whole function of phosphorus in the turkey is discharged. The radioactive measurements thus held the possibility of a much more thorough study of phosphorus metabolism, only part of which is concerned with vitamin D.

This experiment leads to the discussion of an important point which has been revealed by tracer work and which is, perhaps, the most significant single result it has produced. Its discovery was actually made by Schoenheimer by means of stable isotopes, and it is more fully described in the next chapter. Schoenheimer showed that, although the proportion of various amino acids in a metabolizing system may remain sensibly constant, the actual amino acids are continually breaking down and reforming. This lability, in which the atoms concerned with a living system are continually changing while the system itself retains the same nature, is apparently a fundamental property of life. It is evident that the stable part of living materials is not unchanging chemical compounds, but rather some overriding process which requires that, in spite of the ability of amino acids to break down and reform, the proportions remain the same. An analogy is the Congress of the United States, which has the same form and make-up but definitely not the same individuals all the time. The overriding process is the Constitution and the people who wish to live by it.

The same kind of lability seems to hold for many elements, including phosphorus. Therefore in the nutrition experiment just described it is not possible to say that  $P^{32}$  fed in the form of phytic acid one day is behaving like the direct products of phytic acid several days later. From the point of view of the nutrition experiment the initial process is of major interest, and the experiment should be designed to study events as close as possible to the original meal of phytic acid.

Experiments such as this with phosphorus are relatively difficult because of the widespread use of phosphorus by the organism. An element such as iron is easier to study since its primary locale is in the hemoglobin and it can be used to give valuable information as to the way in which iron is assimilated in various kinds of diet. Very valuable studies of this kind have been made by Hahn, Bale, Whipple, and others at the University of Rochester. One simple result seems to be that the uptake of iron is a competitive matter so that iron should be fed to an individual when the stomach is empty.



A second illustration of an attack on a definite problem is the application of tracer methods to the problem of photosynthesis. All that can be said definitely about photosynthesis apart from recent work is that a plant or allied organism, notably green algae, can take  $\text{CO}_2$  and water, plus radiant energy, and convert them into complicated carbohydrates and oxygen. The overall achievement is so sensational and has so little to characterize it that it is not surprising that it has proved to be a baffling problem. The most persistent mechanism postulated was that of Willstätter, who proposed that the process proceeds via formaldehyde, which then polymerizes in some manner. This process served in the absence of a better for some years, even though formaldehyde is toxic and had never been detected in sufficient amounts. A striking advance in understanding this process came when Ruben and Kamen used  $\text{C}^{11}$  to study the fate of radioactive  $\text{CO}_2$  exposed to algae in the presence of light and dark. They were able to show clearly what had been already suspected, that a reversible dark reaction takes place in addition to processes involving light and also that the ability to cause photosynthesis is stored and can take place after light is removed. Moreover, the oxygen formed in the process was shown by the use of  $\text{O}^{18}$  and a mass spectrograph to be derived from water and not in any part from  $\text{CO}_2$ . This eliminates the simple formaldehyde suggestion.

The present situation regarding photosynthesis is far from one of complete understanding, but a great deal of the mystery has been removed by tracer work. It is evident that the process is stepwise, with only part involving light directly. One of the intermediate products seems to be succinic acid. It is also likely that there is no one single process of photosynthesis, but rather varying methods adapted to the particular organism.

One very valuable use of tracer techniques is in studying the so-called "trace elements." Almost any metal is valuable in some way in the living organism. Usually it is needed in minute amounts only, and these are in all likelihood not distributed widely but confined to one particular compound. Therefore rapid progress can be made with the biochemistry of such a compound. A very beautiful illustration of this is the work of Abelson with vitamin  $\text{B}_{12}$ . This remarkable substance is the active principle of liver in the cure of pernicious anemia, and it has the most astonishing effects on the ability of a chick to use its food efficiently, so much so that the future cost of chicken meat should be halved.



This vitamin contains cobalt. By using radioactive cobalt of high specific activity Abelson was able to show that colon bacteria would take up 75 per cent of the radioactive cobalt and form vitamin B<sub>12</sub> containing radioactive cobalt. By means of this the necessary processes for separation of the vitamin were completed very much faster than would normally be possible, so that in a few weeks adequate methods of biosynthesis and chemical separation were worked out. For instance it was shown that *E. coli* will not synthesize vitamin B<sub>12</sub> unless there are traces of iron or calcium in their diet. Very exciting work lies ahead with this radioactive vitamin.

An interesting application of artificial radioactivity to historical time measurement has been made by Libby. It is likely that C<sup>14</sup> is formed from N<sup>14</sup> in the atmosphere by neutrons generated in cosmic rays. The reaction is  $N^{14}(np)C^{14}$ . This C<sup>14</sup> will become incorporated by living systems to an equilibrium amount, but on cessation of metabolism it will diminish in activity on account of decay. A mummy that is 3000 years old will thus have less than normal C<sup>14</sup> activity. This can be measured. The method has considerable promise in archaeological research.

## BIOLOGICAL EFFECTS OF NUCLEAR RADIATIONS AND NEUTRONS

We can now pass on to the rather different subject of the biological effects of nuclear radiations and some of the possible applications of these effects. It is well, at first, to consider what the biologist has deduced about the fundamental nature of living processes and then see what the physicist has to say about the action of radiation on living systems. The unit of living material is the cell. This is complicated but exhibits two striking features which are related. In the cell quite amazing processes take place which are made possible by enzymes or by growth factors such as vitamins. It appears likely that both these are in turn regulated by remarkable entities, called genes, which unquestionably control the ability of the cell to produce the necessary enzymes or growth factors. In each cell there are very few identical genes; in fact it seems likely that for a large part of its life the cell has only one effective gene of each kind. A gene is the size of a very large molecule and has an effective diameter of perhaps 200 Angstrom units, this being a very broad statement. A gene is very stable, at least in its proper environment, and its half-life can be estimated at a million years. Prior to the process of cell



division the gene produces an exactly similar entity by a most intriguing and as yet mysterious process, and after division the function of the gene is continued in both cells. Cells divide at widely different rates, ranging from practically not at all in the crystalline lens of the eye, to every twenty minutes for bacteria.

The reader can at once see how radiation will affect the cell in a general way. Radiation is penetrating so that it has access to the whole body of the cell, something which is not true of chemical agents. Therefore it can affect everything the cell does. However, if it affects the nature of a gene then the whole subsequent behavior of the cell is changed because the enzyme or growth factor content of the cell is altered and therefore the whole delicate balance of mutual competition between different processes is upset. It is not unexpected therefore that the most dramatic effects of radiation are genetic. It is also true that these are not the only effects.

We can now consider what happens to a large molecule (which as physicists we find ourselves using to represent a gene) when nuclear radiations are in its vicinity. In the first place we need to know something about the binding between different parts of the molecular structure of a gene. Evidence for this can be obtained from the action of heat on genes. As the temperature increases, the rate of mutation increases, and the manner of increase fits in with a binding in one place of about 3 electron volts or 69 calories per mole. Now almost anything that radiation is able to do involves a local release of energy well in excess of this. Thus a fast electron, which is to be thought of as a moving intense electric field, can cause local releases of energy of the order of 100 electron volts and certainly is readily capable of producing excitations of one-tenth this size even if it does not produce ionization. A fast neutron can produce a recoil atom in a neighboring molecule, in which case a tremendous swath of ionization which is capable of releasing two or three hundred electron volts inside the gene is let loose. Or the neutron, if slow, can gently attach itself to a nucleus and transmute it to a new nucleus which then reverts to the ground state with the emission of one or more gamma rays totaling 8 Mev in energy. The recoil of these good-sized photons sends the new nucleus reeling backwards with about 50 electron volts of energy, and this is actually the most gentle treatment the gene can ever expect if it suffers a direct encounter.

The apparently unescapable conclusion is therefore that if some direct effect of radiation takes place inside a gene the gene ceases to behave as it used to do. This is the basis of the so-called "target"



theory of the action of radiation. One or two points need to be made about this. The first is that a molecule with a molecular weight of a million is quite an empire in itself and for that reason may prefer to revert to its rightful configuration even though an explosion has taken place in its midst. Put differently, the fields of force binding the molecule are extensive and, if an atom or two are dislodged, they may well be able to lose their excess energy received as a result of the radiation and then return to their original locations. This process is called the "cage effect." Another point is that not all the molecule may be equally important, so that even though damage is incurred in one locale it may not destroy the power of the gene to perform its function and also to recover its proper shape. This is illustrated by the fact that bacterial viruses are vulnerable to effects from deuteron bombardment only in about 40 per cent of their actual volume. A virus has many features in common with a gene, and it is an attractive hypothesis to suppose that viruses are freely existing genes, not attached to any cell.

In default of a complete understanding of the action of radiation the authors are attracted to the target theory. There is one very important additional method of getting at the target, and this is by means of active centers produced by the radiation which are able to move around inside the cell until they encounter the gene and there produce a change. A moment's thought shows that this is not only possible but extremely likely. The energy that radiation could give to some active molecule, like hydrogen peroxide or a free H or OH radical, is in the order of 5 electron volts. The energy necessary to change a gene we have seen to be 3 electron volts. Therefore the collision of such a molecule with a gene will cause a change, and the chance of such a process is quite good compared with that of producing an ionizing event inside a target with a diameter 200 Angstrom units.

What therefore occurs seems to be this. Radiation in some way produces either ionizing particles (electrons for x-rays or gamma rays; protons or recoil nuclei for neutrons), or else a recoil event as described above for the neutron capture process. The fact that neutrons or gamma rays are not readily absorbable means that they themselves produce no effect until they produce secondary action deep inside the material and hence the effects of such radiation are transmitted through the whole volume. These secondary particles then produce either direct effects in the genes themselves, or else produce a high concentration of freely moving energetic centers (such as H or



OH radicals) which collide with large molecules and therefore occasionally with the genes. Either of these events then produces a lasting effect on the cell which may manifest itself in various ways, the most likely being a reduced ability to divide. An attractive theory for cancer is to suppose that such a process has taken place in a somatic cell (i.e., one which is not concerned with the reproductive process) and that by bad luck the alteration of the gene has been such as to produce a cell which is lacking in the factors which stop growth at the time when the whole organism calls for it. The growth is therefore wild and unchecked, though not in itself toxic, until by sheer size it encroaches on some bodily function.

To fit in with this theory of the action of radiation there has recently been discovered a series of chemical agents which are capable of producing gene changes or mutations. These are to be likened to the indirect method of access to the gene.

The reader may by now be impatient at the preoccupation with the genes and wonder what happens to the rest of the cell which is so much more extensive. The answer is that non-genetic effect does indeed take place, and in a large organism like man, where the structure of cells is all settled and a few cells destroyed by genetic action can be dispensed with without harm, the effect of radiation on cellular material can be most serious. What occurs then is a species of poisoning of an internal character, due to the release inside the cell of breakdown products which are not necessary to the living process. In addition there may be a starvation due to the destruction of enzymes. This is a very broad description of radiation sickness, but it shows that there is hope for alleviation of a sufferer, particularly if some critical poison is counteracted enough to enable the individual to survive and exercise his adaptive powers. Some progress along these lines has been reported.

Returning to the target theory for a moment, it is worth mentioning that by applying essentially the reasoning of p. 106 it has proved possible to estimate the cross section of various targets. This has especially been exploited by Lea. The method used relies on the ability of self-duplicating organisms to make sufficient of their like to show their presence. Thus colonies of bacteria can be grown from a single bacterium, and the number of colonies developing is a measure of the bacteria originally present. By irradiating the bacteria and determining the number which survive, the size of the target which has to be hit can be determined. The measurements so far are not extremely accurate because allowance for indirect effects has to be made



very carefully. Nevertheless the figures are yielding very interesting information on the physical size of self-duplicating entities.

The controlled use of radiation for biological effect is in its infancy. The reason is that up to the present the majority of the work has been done with the most readily available radiation—x-rays—and these exert a high-energy, non-selective effect. The remarkable fact that tumor tissue is more susceptible to radiation than normal tissue has been used with remarkable results, and it can be claimed that apart from surgery this is the only method of cancer cure. Nevertheless, consider the possibilities ahead if slow neutrons are used for irradiation. These are of themselves no more likely to produce damage than hydrogen or helium molecules because they have no more than thermal energy. On the other hand the nuclear absorption of these is to some extent controllable by making use of nuclear resonances and by picking on a particular element for selective action. A start on this was made by Kruger. He irradiated tumor tissue in boric acid solution with slow neutrons, hoping that the energy released by the slow neutron reaction with boron (which releases an alpha particle and a lithium nucleus) would give a preferential action on the tumor. Some success was had. At that time the only source of slow neutrons was from a cyclotron, and the presence of fast neutrons was unavoidable. The thermal column of a pile is far more suitable. We predict very important results from the combination of slow neutron bombardments with biochemistry and biology.

### Dosage measurements for radiation

Dosage measurements of radiation are difficult and unpleasant and will get increasingly more so. The reason is the wide variety in the action of radiation, both on account of the nature of the radiation itself and of the material on which it acts. Radiation measurement therefore resembles somewhat a subject like economics, in which one can watch the stock market, or the business index, or the number of business failures, each of which tell some of the story of the economic situation, yet not all of it.

Present-day dosage measurement centers around a radiological unit, the *roentgen*. The roentgen is a total dose, not time dependent, and it is measured by ionization under circumstances where the radiation is fully in equilibrium as regards the production of ionization. When this condition obtains, 1 roentgen is that amount of radiation which will produce one electrostatic unit of charge as ions in 1 cc of air at 0° C and 760-mm pressure. When this is properly reduced to energy figures



1 roentgen is 83.8 ergs released as ionization per gram of air. To measure this, thimble ionization meters have been devised in which the material of the thimble is such that equilibrium between primary action and secondary ionization is practically the same as in a large volume of air. Since such meters are convenient, roentgens are commonly quoted. Such measurements should always be accompanied by an account of how they are made.

Since roentgens are measured by definition in air an extension of the definition to the effect in tissue has been proposed. A *roentgen-equivalent-physical*, or rep, is defined as radiation producing 83.8 ergs per gram of tissue. Actually this is a new method of measurement, for Spiers has shown that the number of ergs per roentgen per gram of tissue varies over wide values ranging from 42 for fat to 880 for bone for soft x-rays.

For neutrons a similar measurement, again based on the convenience of the thimble ionization chamber, can be made. One  $n$  is the number of neutrons which will produce the same action on a Victoreen thimble chamber as 1 roentgen of gamma radiation. In terms of grams of tissue this is 190 ergs.

Continuing further with indices of radiation there is the need for an index of biological action. This is a wide-open subject, depending acutely on the method of measurement. The roentgens needed to produce the same effect as standard gamma radiation are sometimes called the roentgen-equivalent-man (rem). Thus in terms of the production of cataract in the eye lens a proposed figure for neutrons is  $\frac{1}{20}n$  equals 1 rem.

### **Relative biological effectiveness of neutrons and x-rays**

The subject of indices for biological action brings up the very important subject of the relative effect of neutrons and x- or gamma radiation. Several methods of estimation have been tried. One is the threshold dose to produce "sunburn" or erythema. One thousand roentgens will produce this, whereas the same reaction is produced by  $200n$  of neutrons. On the other hand, genetic effects are more readily produced by neutrons and a factor closer to 10 than the 5 given above is indicated. The latest (and incomplete) information concerns lens cataract for which the ratio appears to be about 20. Neutrons must therefore be treated seriously and low tolerance figures adopted.

### **Protection of workers in radiation laboratories**

The general interest in the protection of workers in the field of nuclear physics from the radiations which they are liable to absorb is



great enough to warrant a short discussion of the matter here. Before treating the more subtle influences of neutrons and gamma radiation, for example, a word on electrical shock is not amiss. Even though it may be boasted that an installation for nuclear research has no exposed high voltage, it is well to remember that the nuclear physicist has a rather specialized idea of the meaning of "high" voltage and is prone to discount voltages of the order of a thousand or two. The ordinary Geiger counter, working at about 800 volts, packs considerable "wallop" and should be understood by all who are using it. Such matters as the accelerating voltage on the filament emission of a cyclotron are often forgotten by the physicist who put in the installation, and yet they may be capable of great, if not final, damage. The fatalities among workers with high-voltage equipment are not many, but they are not zero, and death due to relatively low voltage has occurred. This point about the relative importance of the accident hazard has been well brought out in a paper by Aebersold on the subject of protection for cyclotron workers, and we wish to add our weight to his remarks.

The effect of radiations is rather more sinister and for that reason is discussed much more than straight accidents. The horrible suffering of early workers with x-rays, who unthinkingly took large doses in quite trivial ways—for example, in showing the "shadow" of the bones of their hand at a demonstration—has made the pioneers in work with neutrons rather more careful, and there is no real reason to repeat any such "martyrdom to science." It is accepted among radiologists that the maximum safe dose in a normal week's work is  $\frac{1}{4}$  roentgen. The safe dose for neutrons on the same basis is  $\frac{1}{80}n$ . The authors take the attitude that these figures represent the safe dose for an individual who does not fear any genetic changes which may be caused by radiation.

Accepting the two "tolerances" given, it is necessary to consider the way in which precautions should be taken. Consider x-rays first. Very roughly, we may say that  $\frac{1}{10}$  roentgen will require  $10^8$  quanta to pass a square centimeter of surface. If this number is all that is given by the machine at a given distance in the proper time then it is safe without any protection. If this is not the case, then it is necessary to interpose some kind of absorber which will reduce the number to the safe figure. This is quite easy for soft x-rays produced by electrons of energy less than 10,000 volts but is a much harder matter for the x-rays now being used in many hospitals where the energy of the bombarding electrons is a million electron volts or more. To reduce the primary intensity of such x-rays by a factor of a hundred would



require about 3 inches of lead, which means a very expensive protecting wall. It is cheaper to interpose a foot or more of concrete between the target in the x-ray machine and the operator. We do not intend to discuss the details of safety precautions for x-ray workers, as they are best studied in the publications of the National Bureau of Standards.

Radiation protection in general is now a subject of wide interest. The first thing to say is that any kind of radiation protection must be checked by measurement. For this purpose radiation survey meters have been devised and are available on the market. Our own procedure is to use a Lauritsen electroscope which has a thin aluminum ionization chamber. Measurements of the ionization are taken with the chamber bare, and with a silver cylinder slipped over the chamber and surrounded by paraffin. An increase in the latter case is due to radioactivity induced in the silver by slow neutrons. From these measurements some idea of the actual radiation hazard can be inferred and also the proportion of the radiation which is due to neutrons. The electroscope can be calibrated in terms of gamma radiation by means of a source of  $\text{Co}^{60}$  which produces 1.3 milliroentgens hr. at a meter distance per millicurie.

Shielding for gamma radiation can be calculated by using the absorption coefficients given in Appendix 5. First there has to be an estimate of the primary radiation. This must be done in terms of expected performance before any new machine is turned on, and the shielding prescribed for the purpose installed before the machine is operated. We stress this because 90 per cent of the danger to personnel occurs when the machine is turned on, and the number of radiation-producing machines being used in the world today is in the hundreds, if not thousands. Each one is a potential source of physical handicap for the operator who is installing the machine. The shielding should then be tested as the radiation is turned on and added to if necessary before final operation is permitted.

Shielding for neutrons is not so easy to calculate because it depends markedly on the proportion of fast neutrons, and how fast they are. Neutrons are absorbed by *nucleons*, that is by the neutrons and protons in nuclei. Everything else being equal, it pays to spread these nucleons out as much as possible so that the lighter the element used for absorption the fewer nuclei are needed to absorb neutrons. Unfortunately everything else is not equal, for the cross section of different nuclei for neutrons varies considerably. There is also an energy dependence of cross section. If one uses for computation the value of the nuclear cross section as determined from the nuclear radius then



the thickness of concrete to reduce the neutron intensity by a factor of 10 is roughly 10 inches. Concrete is a reasonable material for shielding, and a thickness based on this figure will not be very far wrong, particularly if distance is also interposed between the neutron source and the operators.

Usually the problem of lateral shielding is not so serious because concrete walls are no great problem. However, shielding must also be applied above and below. Shielding below is also no problem because not many accelerators are light in weight and there are not often personnel below such a machine. For shielding above the thickness of shield can be reduced by using alternate layers of iron and paraffin or concrete. The iron has a relatively high cross section for inelastic scattering of neutrons so that the energy of the neutrons is rapidly reduced and they become more readily absorbed in the paraffin. Such absorption is actually only a matter of exchange because the act of absorption of a neutron also produces a gamma ray, unless the absorption is in boron or lithium where particles are given off and no radiation results. Except that it is expensive, borax is a useful component of a shield.

All radiation is scattered much more than seems likely at first. Therefore there should be only narrow openings, which are themselves shielded, through the shield and extra care should be taken in measurements to check that no scattered radiation is in unwanted places.

To sum up the situation on radiation protection, we can say that for most purposes a dose rate of 250 milliroentgens per week total is satisfactory provided there is no neutron content. The neutron-produced part of this ionization must be given an extra factor of 20, so that the principal shielding problem is concerned with neutrons.

The reasons for shielding are primarily to avoid unknown factors, such as lens cataract, and genetic effects which have already been described. It is our philosophy that no one should be exposed to more than five times the cosmic-ray dosage as a total over a period of any one year. This is restrictive but probably wise.

To keep track of radiation received, film badges are very useful, and for each day when exposure is known to be possible, pocket ionization chambers should be worn. These are used in connection with an electrometer and show the total ionization received in a small ionization chamber during any period. They have the advantage that there is no waiting for processing. Two should be worn as the indications from one alone are not always reliable and confirmation of a heavy dose by a second chamber is worth having.



Five hundred roentgens over the whole body is a fatal dose. This is worth remembering when a new machine is being turned on.

We can conclude with a word about beta rays. These are similar in action to the secondary electrons produced by gamma rays except that their action is confined to a rather thin surface layer. It is very intense, about thirty times more so than gamma radiation itself. It is therefore important to avoid even short exposures to primary beta rays, for a minute's exposure to the beta rays from 100 millicuries of  $P^{32}$  will produce a definite effect on the skin. This caution is even more significant for alpha rays or, still worse, the direct beam from the cyclotron or other accelerator. A few cases are known in which the direct beam has been taken by an individual on the hand, and in each of these it was fortunate that the exposure was only a matter of seconds, for the burns produced take months to heal. Where remote control is essential, as for a large cyclotron, it is important to ensure that no person can be within range of the beam while the machine is on.

## Outlook

We have made a practice of summarizing at the end of the previous chapters. It seems more fitting here to substitute a short section in which we discuss the future for the application of nuclear physics. We are thus for the moment in the role of prophets, and we do not claim that we are necessarily right.

We look to the most significant results from the field of tracer work in biology, botany, and their attendant sciences. Where the tracer work is carried out among chemical compounds which do not freely exchange their elements, clean, accurate results should be found once a few techniques have become standardized. The amount of literature already accumulating testifies to the safety of our prediction. We look to a rich, but rather slower, development of radioactive tracer work in chemistry proper, for accurate quantitative results are important in that field. This means the development of reliable measurement methods and will take time. The chemists are still feeling out the new technique, and obvious applications are not being found so fast. The same is true of metallurgy.

As far as therapeutic work is concerned it is hard to predict. One field which will take years to develop, namely, the use of tracers in diagnosis, seems attractive, but it is foolish to say that there is a great future for a nearly untried method. It is quite likely that artificial radioelements will take their place beside radium for the kind of treat-



ments given today. The future of neutron therapy still awaits further work before we can make any statement.

The last two paragraphs lead up very nicely to the one general field which is certain to be of interest, and that is research in the whole subject. The great pleasure we derive from writing this book stems from the fact that we are able to describe the weapons to be used in an attack on one of the strongholds of nature and are also able to be among those actually engaged in battering at the stronghold. It is a new subject, not one in which the dotting of i's is all that remains. Rich prizes are yet to be won in this domain of science, perhaps without extreme effort, and it is the sense of pioneering and possible rewards which keeps the interest stimulated.

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## 9. Stable Isotopes and Their Application

### Isotopes in general

In the first chapter, and often in subsequent chapters, we have spoken of the building blocks of nature as neutrons, protons, and electrons. The neutrons and protons were the constituents of the atomic nucleus, and the electrons made up the outer atom, their number being just sufficient to render the whole atom neutral. In these chapters we have pointed out many times that it is possible to have nuclei which are off the beaten track, with an unusual number of neutrons relative to the number generally found. It has been implied that such nuclei are radioactive, and as a first step in understanding radioactivity such an implication is fair enough. In actual fact, however, very many atomic nuclei having a certain definite number of protons, which number characterizes the element, can contain several different numbers of neutrons and yet remain stable. The discovery of this fact has been of the utmost importance in nuclear physics. It was first suspected when certain of the naturally radioactive elements which differed greatly from one another in their radioactive properties were found to be chemically inseparable, suggesting that their external structures are identical though their nuclei differ. This early suggestion did not, however, specify anything about the possibility of *stable* elements existing in forms which differed only in the nuclei, and it was not until the celebrated parabola experiment of J. J. Thomson in 1912 that it was shown that neon existed in two forms.

This experiment is worth a little consideration, even though rather different techniques are used today, because it shows rather well the fundamental problems of detecting isotopes. The main object of the experiment is to associate a charge with a nucleus and to subject it, in this charged condition, to the action of electric and magnetic fields in such a way that they cause deflections of the moving charged nuclei



which depend on the mass of the nuclei. If nuclei were normally supplied with no attendant electrons the problem of associating a charge with them would clearly not arise as the nucleus is already charged.

In actual fact the only available form in which nuclei can be obtained is as part of a neutral atom, and it is therefore necessary to ionize the atom in order to produce the associated charge. This can be done quite easily by several methods, that chosen by J. J. Thomson being the discharge. A discharge was produced in a large glass dis-

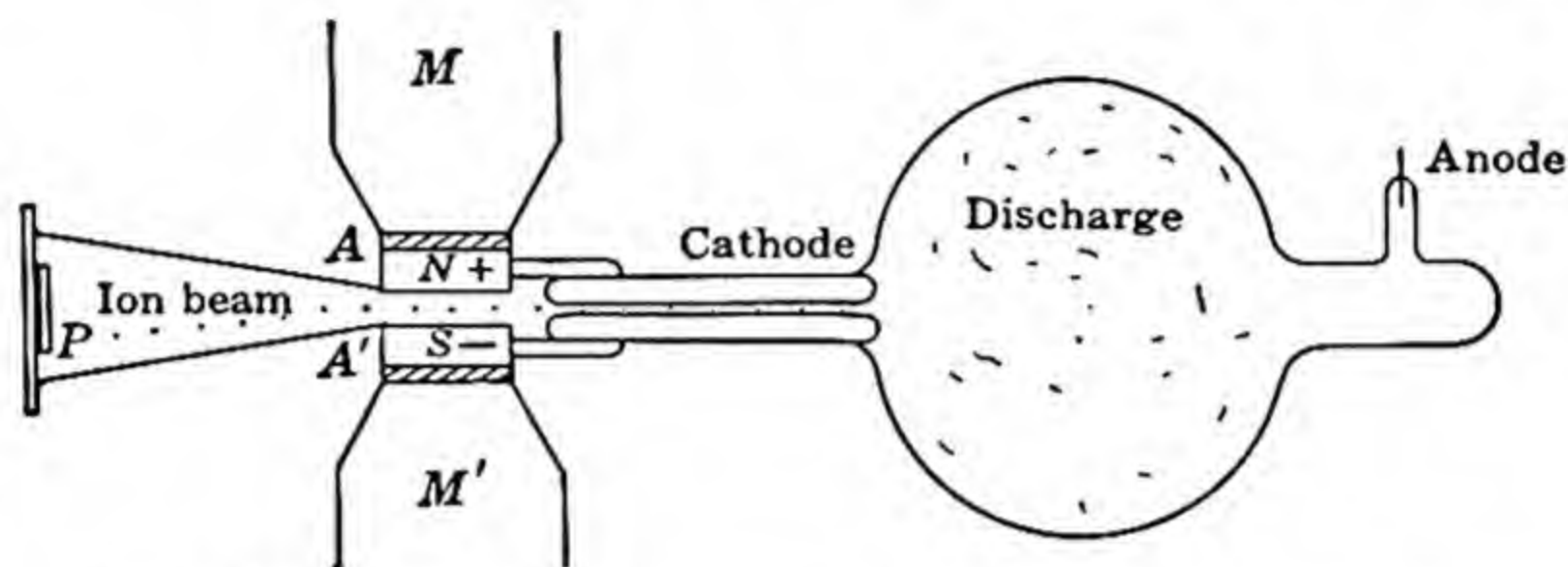


FIG. 1. Essentials of J. J. Thomson's parabola method of studying positive ions. The ions are formed in the discharge tube. A few of them pass through the hole in the cathode and into a space (N, S; +, -) where they are subjected to parallel magnetic and electric fields. These produce deflections at right angles to one another, and traces in parabolic form are found on the photographic plate. Ions having different charge-to-mass ratios produce different parabolas, so that two parabolas from one element constitute evidence for the existence of isotopes.

charge tube, using rather high potentials; and by boring a hole in the cathode it was possible to obtain a fine jet of positive ions. These ions were traveling at high speeds, and owing to their manner of formation their range of speeds was considerable. The ions were then made to pass into a space in which they could be subjected to the influence of parallel electric and magnetic fields.

In Fig. 1 the apparatus is represented schematically, and the poles NS of the magnet, and the two electrodes + and -, were composed of the same two pieces of iron, insulated from the remainder of the magnet MM' as shown. The charged ions are subject to two different forces in this space AA', forces which act perpendicularly to the motion of the ions and also mutually perpendicularly. The magnetic field alone causes a force to act which is equal to  $Hev/c$ , where  $H$  is the magnetic field intensity;  $e$  the electrostatic charge carried by the ion, and  $v$  the velocity of the ion. This force acts perpendicularly to the path of the ion and so produces an acceleration of constant magnitude perpendicular to the motion. This means that the motion is circular



with a radius of curvature  $r$  such that the mass  $m$  times the acceleration  $v^2/r$  is equal to the force causing the acceleration. That is,

$$\frac{Hev}{c} = \frac{mv^2}{r}$$

or

$$r = \frac{mvc}{He}$$

Now a little simple geometry will show that the deflection from the original path taken by a neutral ion will be inversely proportional to the radius of curvature, at least to the first approximation, and so we have for the magnetic deflection  $d_H$ , where

$$d_H = A \frac{He}{mvc}$$

$A$  being a constant. The ions will be spread out into a streak, the streakiness being due to the non-uniformity of  $v$ . All such streaks will be superposed no matter whether  $m$  varies or not. However, if the electric field is now turned on, these streaks will be pulled out to one side. If the manner of deflection were just the same it would not help, for it would simply turn the streaks around. But the manner of deflection is different, for the force does not depend on the speed of the particle, but only on its charge and the intensity of the electric field, which we can denote by  $X$ . The force is then  $Xe$ , and this produces an acceleration  $a$  given by

$$Xe = ma$$

The ion then "falls" in this field in the manner of a projectile while between the electrodes, and so is deflected a distance  $s$ , where

$$s = \frac{1}{2}at^2$$

$t$  being the time spent between the electrodes. This time, however, is the length of the electrodes  $l$  divided by the velocity  $v$ , so that

$$s = \frac{\frac{1}{2}al^2}{v^2}$$

or

$$s = \frac{\frac{1}{2}Xel^2}{mv^2}$$

where we have substituted  $Xe/m$  for  $a$ . Again, by simple geometric reasoning it can be shown that the deflection  $d_X$  due to the electric field is a constant  $B$  times this, or



$$d_X = B \frac{\frac{1}{2} X e l^2}{m v^2}$$

Now, if the values of  $H$  and  $X$  do not change, each ion having a definite value of  $e$ ,  $m$ , and  $v$  will have a definite place on the photographic plate, and if there is a continuous spread in the values of  $v$  those ions having the same  $e/m$  will lie along a definite trace and those having a different  $e/m$  will have a *trace separated from the other*. This is easily seen in terms of the equations by eliminating  $v$  from the two expressions for the deflections and then getting

$$d_X = \frac{X l^2 B c^2}{2 A^2 H^2} \cdot \frac{m}{e} : d_H^2$$

which shows that the traces are parabolas but different parabolas for particles with different  $m/e$ .

Thomson's experiment showed many parabolas which could easily be explained as due to molecular ions or multiply charged ions, but in neon, where no molecules are formed and the gas is light, two traces were always found close together, and no matter how the discharge was run the relative intensities of the two were always the same. No explanation could be found for this fact other than the existence of two forms of the one element neon, the difference being only in the mass of the nucleus of the atom, as the chemical properties (and hence electronic structure) were the same. In terms of our present-day knowledge of the constituents of nuclei we say at once that the nucleus of the light form has ten protons and ten neutrons while the heavy form has ten protons and twelve neutrons. The name *isotopes* was given to such nearly identical atoms.

We propose to return to the point later, but we wish to point out here that the experiment showed that the ratio of the intensities of the two isotopes remained the same under all conditions. This is an essential feature of all normally occurring material; the ratio of the concentrations of the various isotopes is always the same.

This discovery of the existence of stable isotopes was followed by the steady uniformly directed work of Aston, who showed the generality of the phenomenon and in the years 1919 to 1925 discovered new isotopes in an almost continual flow. For his experiments the fundamental idea of the parabola method, to subject positive ions to electric and magnetic fields, was retained, but the method of subjecting the ions to these fields was changed to give better resolution among different isotopes. The main defect of the parabola method is that it does



not tend to focus the ion beam. Aston directed his attention to a method by which ions which had different velocities but the same  $e/m$  would arrive at the same point on the photographic plate *even though they started their paths at different angles*. This is achieved by separating the electric and magnetic fields so that the electric deflection is applied first and the magnetic afterward. The two deflections are also arranged to be in the same plane as the plane of the beam; that is, the electric and magnetic fields are perpendicular to one another. Now the reader can see from the equations for the parabola method that the electric deflection of a fast ion is small, depending on  $1/v^2$ , whereas the magnetic deflection depends on  $1/v$ . By letting the ions travel a long way after the influence of the electric field and only a short way after the influence of the magnetic field, and by choosing a suitable inclination for the detecting plate, it is possible to obtain a beam of ions, focused at one point on the plate for variable velocity and angle of origin, but the *same*  $e/m$ . This property of focusing mass is similar to the action of the spectrograph in focusing light of various wavelengths, hence the term "mass spectrograph" or "mass spectrometer."

From the point of view of nuclear physics some of the most interesting results have been obtained with mass spectrographs of very high resolution; but, as such work is highly specialized and the apparatus elaborate, we prefer here to describe only mass spectrometers which are suitable for analysis of elements into their constituent isotopes, and hence of interest in the application of isotopes to organic chemistry and biology.

### Mass Spectrographs

The problem which dominated the early experiments on isotopes—devising an apparatus which would focus a beam of ions having widely different velocities—is now no longer the major difficulty. One very simple method was suggested and put into practice by Dempster between 1918 and 1922. This method utilizes the fact that particles bent in a semicircle will be focused at the end of their path and so may be detected with accuracy. The best way of seeing this is to construct circles of the same radius but with centers a little above one another. It will be seen that though the circles can be far apart above the centers they lie quite close together near the horizontal diameter. This focusing action is often made use of in nuclear physics, for example in beta-ray spectrometers. In Dempster's experiments the positive ions were produced by bombarding atoms of an element evaporated from a hot surface, by electrons accelerated after leaving a hot tungsten



wire. Such ions are formed with very little energy other than their energy of ionization, and if they are then made to fall through a definite potential difference of about a thousand volts they will each have virtually the same energy. After this they are made to pass through a slit system in a magnetic field and enter a chamber where they are collected on an insulated plate and their number measured as a current on an electrometer. The magnetic field will not be of the right value to bend (and hence focus through the slit system) the ions into semicircles unless it is specially adjusted, and each isotope will have its own particular value of the field. By plotting the detector current against magnetic field the various isotopes appear as peaks of current, and their abundance may be compared by comparing the heights of the peaks.

A second method of obtaining a definite velocity for the ions is that used by Bainbridge. Bainbridge subjected the ions to a preliminary passage through a region in which electric and magnetic fields applied opposing forces. If the forces are equal the ions can pass through the slit system, and as this equality requires that

$$\frac{Hev}{c} = Xe$$

the velocity  $v$  is determined by the ratio of the electric and magnetic fields. These ions that have passed through the "velocity selector" are then bent into semicircles and detected on a photographic plate. Since the velocity is fixed the various semicircles can correspond only to definite values of  $e/m$ .

The design of mass spectrometers for routine work with separated isotopes is becoming standardized. Two designs have found favor in laboratories where separated isotopes are being used, the designs of Nier and Bleakney. As Nier's design is more recent and quite simple it will be described first.

The apparatus does not look particularly like Dempster's arrangement, yet it differs in principle only in the focusing arrangements. The ions are produced by bombarding by electrons the gas or vapor to be studied. These ions are accelerated by a potential drop of about a thousand volts, and the beam of ions so produced passes into a magnetic field in which they suffer a deflection of  $60^\circ$ , rather than  $180^\circ$  as in Dempster's method. This deflection permits the use of a very simple magnet and succeeds in focusing the ions owing to a fact first pointed out by Barber, that, if a magnet having pole pieces in a wedge shape is



used to deflect ions and the ions enter and leave the pole pieces perpendicularly to them, then the source slit, the apex of the wedge, and the focus lie on a straight line.

The experimental arrangement is as indicated in Fig. 2. The ions are formed by electron bombardment from the filament  $F$ , in the space indicated by the dotted lines.  $C$  is a wire attached to the collecting electrode used to measure the bombarding current. The ions are accelerated to the slit in the plate  $P$  by a variable potential of around

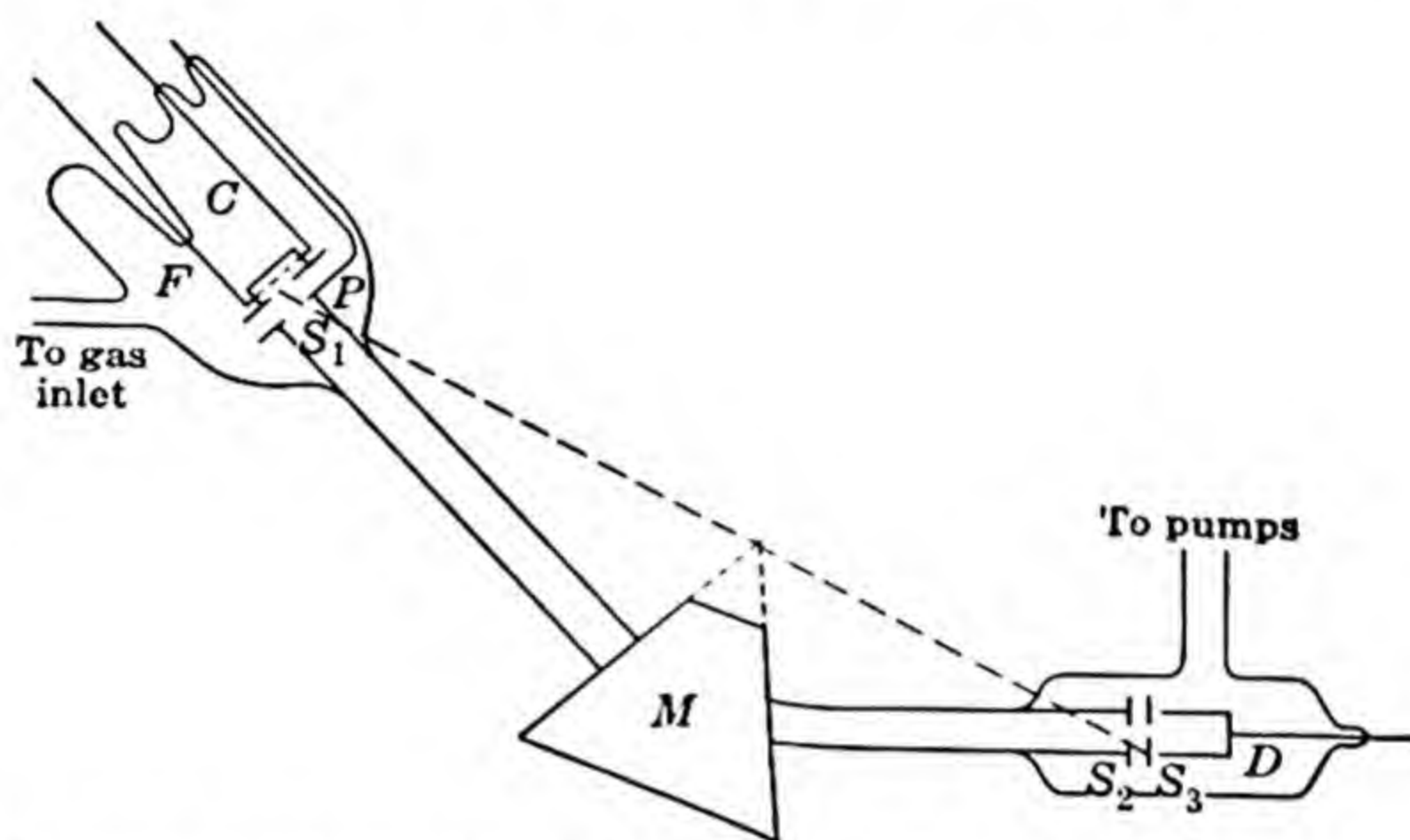


FIG. 2. Nier's mass spectrometer. The ions are formed by bombardment of electrons in the space indicated by dotted lines. They are accelerated by a variable voltage to the slit  $S_1$ , pass down the tube to the magnetic field  $M$ , and are focused by it on the slits  $S_2$ ,  $S_3$ . The slit  $S_3$  is used to apply a repelling potential to any secondary electrons formed at  $S_2$ . The ion beam is detected by an electrometer tube.

a thousand volts and proceed down the tube to  $M$ , the magnetic field. Here they are deflected and then are focused on the slits  $S_2$ ,  $S_3$ . The second slit  $S_3$  is needed to apply a repelling field to keep electrons produced in the first slit  $S_2$  by positive-ion bombardment from registering in the recording equipment. These secondary electrons depend on a number of factors, and although at any given time the number of secondary electrons is proportional to the positive ions their number may vary as the bombarding energy is changed and so interfere with accurate abundance figures. The very small current of ions at the collector is measured by an electrometer tube arrangement as described in Chapter 3. The slits are so arranged that the peaks have flat tops, which greatly helps in making abundance measurements.

To focus different isotopes at the collector the accelerating potential is varied. The relation which governs the curvature of the ion paths is the familiar relation



$$\frac{Hev}{c} = \frac{mv^2}{r}$$

and the velocity of the ions is given by

$$\frac{1}{2}mv^2 = Ve$$

where  $V$  and  $e$  are in electrostatic units in the second equation and  $H$  and  $e$  in electromagnetic units in the first. If  $V$  is expressed in electron volts,  $H$  in gauss, and  $m/e$  in atomic mass units divided by electronic charge, the formula which applies is

$$\frac{m}{e} = 4.82 \times 10^{-5} \frac{r^2 H^2}{V}$$

Then, if the value of  $V$  is changed, with  $H$  fixed, the various values of  $m/e$  will fit the focusing conditions and register on the meter. According to Nier a current of  $5 \times 10^{-11}$  ampere is readily detectable without much difficulty, and it is possible to carry out an analysis with 1 cc of gas at 1-cm mercury pressure, which is about 1 microgram of material. The cost of the whole equipment, if it is built and set working by the laboratory, is about \$1800.

Bleakney's mass spectrograph is also commonly used. It is of the conventional Dempster design, the most important feature being that the whole evacuated space in which the ions move is of glass, which makes it extremely simple to bake out and so remove occluded gas from the solid parts. The magnetic field is produced by air-cored coils and is rather expensive to construct.

A few technical points are of interest. First is the ability to heat the whole apparatus to remove trapped vapors, the process known as "baking out." This is achieved in Nier's apparatus by rolling the magnet out of the way and replacing it by a furnace which heats the copper pipe while the pumps are running. A second point is the presence of "natural" impurities. Both carbon monoxide and water vapor can be expected to show up every time and must be allowed for. Nier suggests that by varying the speed of the electron bombardment in the space where the ions are formed it would be possible to diminish the proportional effect of impurities. Thus OH requires a higher ionization potential than CH<sub>4</sub>, and by means of a rather low-energy bombardment the OH line might be reduced to a minimum.

During World War II the development of pulsed techniques for radar and other purposes led to the suggestion that pulses of ions can be formed and set in flight and the various isotopes detected by the



time of flight along a path a meter or two long. Such an instrument has been made to work, but it seems doubtful whether it is sufficiently simple to replace the magnetic method. For the assay of deuterium it might be feasible since the ratio of masses is so great, and it is possible that an instrument designed primarily for this purpose may find future application. A combination of this idea and a magnetically guided helical path is being tried by Goudsmit and has considerable promise.

The subject of the determination of deuterium is of some interest since it can be made with relatively simple instruments. The density of heavy water is so much greater than that of light water and the operation of comparing density is so accurate that deuterium can be estimated to 1 part in 300,000 by this method. There is one point which must be remembered and that is that the difference in mass of the two hydrogen isotopes is so great that there is actually a definite modification of the electronic structure. As a result the use of deuterium as a tracer may be open to some question. This can be guarded against by careful check experiments. The method for density determination consists of accurate timing of a drop of fluid to be tested falling through a fluid of known density with which it will not mix. The fluid must be carefully kept at constant temperature.

### Separation of isotopes

We have so far explained the discovery of isotopes and the method of detecting them. This does not explain their application in other studies. The reason for their usefulness is that nature has arranged matters so that there is almost no deviation from the abundance ratios of various isotopes, no matter in what form they are found. Such deviations as are found are of the order of one part in a million, with a few exceptions in which the isotopes may result from radioactive changes. This fact means that a sample of material which has an abnormal ratio of isotopes, though it is chemically identical with ordinary material, is nevertheless detectable by the mass spectrometer. Thus, if carbon dioxide containing 10 per cent  $C^{13}$ , in place of the usual 1 per cent, is given to a plant to breathe, any part of the plant which subsequently shows that the proportion of  $C^{13}$  is abnormally high must have received that  $C^{13}$  from the labeled carbon dioxide. In this way the use of separated isotopes is similar to the use of radioactive isotopes, the only difference in principle being that the mass spectrograph acts as a "counter" which will detect non-radioactive material. This being so, the problem of the biologist or organic chemist who expects to



use isotopes as tracers is to obtain elements which have been enriched in one or more of their isotopes. This requires consideration of the process of separating isotopes.

Clearly the problem of isotope separation is not simple, except for two freak materials, deuterium and light helium, where unusual methods can be employed. Leaving these two aside for the moment, it can be seen that the very small difference between elements which differ only by a neutron more or less in the nucleus is going to be exceedingly hard to utilize in separation. Nevertheless, if the amount of material required is not large it should be possible to make use of these small differences and secure some kind of separation if one is ready to make an apparatus elaborate enough and have enough patience. Many such elaborate arrangements have been tried, but at the present time only two are of much use in practice, the method of thermal diffusion and the method of chemical exchange.

Before describing these methods more fully a word about isotope separation in general is in place. In ordinary material we have a mixture of components which we seek to separate one from the other to give us material with enriched isotopes. This is not unlike the problem of obtaining energetic and "cool" molecules from a mixture of the two at ordinary temperatures, a problem which, as is well known, requires the use of some external agent doing work to produce the required result. In thermodynamic terms we are seeking to reduce the entropy of the substance, and we can do this only by the expenditure of the appropriate amount of energy. The hope that some extremely ingenious inexpensive method of separating isotopes is about to be devised is therefore doomed. We should rather expect that all the various possible ways of isotope separation would tend, when efficiently used, to be of about equal value.

With this preliminary word let us consider first the method of thermal diffusion, which can be discussed more generally than chemical exchange. The explanation of this method in easy terms seems to be difficult. In brief, it is the combination of two factors: the first, the factor of thermal diffusion which in many gases causes a concentration of a lighter component near the hotter of two surfaces; and the second, the factor of convection which can be used to "cascade" the separation produced by the process of thermal diffusion. Thermal-diffusion equipment therefore consists of long tubes, cooled on the outside, with a hot wire, or cylinder, along the inner axis. The gas is allowed to remain in these tubes for some time until the two processes result in a separation of the heavy component at the bottom of the columns and



the light at the top. The separation factor depends on many variables, mainly on the length of the column, the temperature difference between the hot wire and the cooled outer wall, and the nature of the gas—the more noble the better.

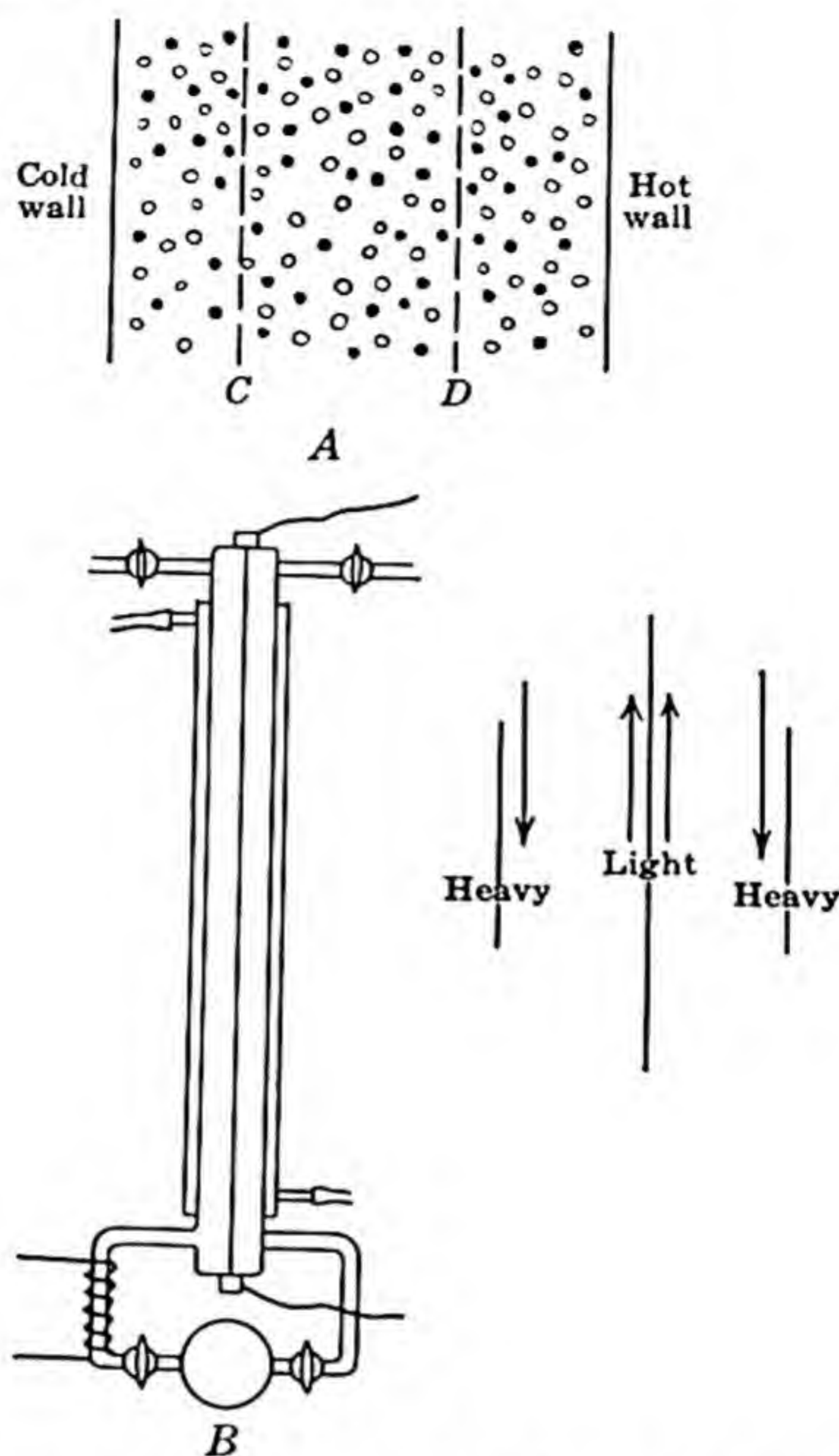


FIG. 3. Schematic representation of the thermal-diffusion method of isotope separation. *A* illustrates the phenomenon of thermal diffusion; *B* is a diagram of a single-column apparatus.

This rather brief account can be amplified by considering the two parts of Fig. 3. In *A* is an attempt to explain the existence of thermal diffusion. The space between the dotted lines, the average part of the gas, can be looked on as separating the regions near the hot and cold walls. Now these regions are still very thick as far as numbers of collisions are concerned, and a faster molecule, after leaving the hot wall, will still have to share its momentum with the molecules in the space near the hot wall before it, or one it has struck, reaches the center. The molecules which arrive at the region *CD* with greatest speeds, and thus with the greatest tendency to diffuse through the layer *CD*, will be



those which have received the greatest amount of momentum from the other molecules in the region near the hot wall. The key to the phenomenon of thermal diffusion is therefore to be sought in the transfer of momentum from one molecule to another, and this will not be an easy matter to predict. In this respect the phenomenon of thermal diffusion brings out clearly the essential complexity of a complete theory of a gas, for the interaction between molecules, which is often dismissed as "elastic," for example, here plays the most important part. It is far from obvious, but it turns out that the transfer of momentum is predominantly from light to heavy for interactions that are very rapidly varying with distance—the so-called "hard-spheres" case—and for an inverse fifth-power law of force disappears entirely. Gases which approximate to hard spheres are therefore easy to separate, whereas those in which the force varies more slowly are difficult. In the first category fall the noble gases; in the second, gases like hydrochloric acid.

Granted, then, that a thermal-diffusion coefficient exists, it will cause a concentration gradient in the gas which will increase until it is balanced by the ordinary diffusion process tending to make the concentration uniform. A single arrangement like *A* in Fig. 3 will thus produce only a very small separation factor, and it is necessary to repeat the process many times to obtain useful separations. The beauty of the thermal-diffusion process is the great ease with which this repetition can be achieved, as can be seen in part *B* of Fig. 3. Here a schematic arrangement of a single column is illustrated. The central hot wire is heated electrically, while the outer part is cooled by a steady flow of water. The thermal diffusion causes the concentration of light isotope near the hot wire and then the ordinary process of convection carries the light isotope upward. This means that as time goes by there will be a concentration gradient not only across the tube but also up the wire, since the gas near the wire is continually being replaced by the convectively carried light component. The result is that the gas at the top of the column is light and that at the bottom heavy. The heavy component can easily be removed by continuing the convective circulation through the flask by means of a heating coil around one of the glass tubes, and after a suitable time the stopcocks can be closed and the flask removed. The light component can be removed similarly.

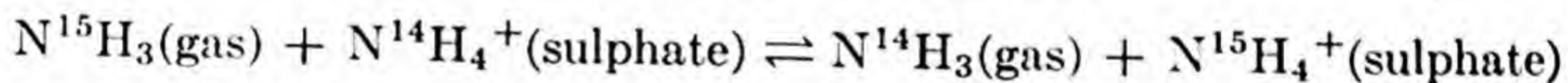
When the gas to be separated is suitable the method is very powerful. For example, Watson found that a single 2-meter column operated



on neon for a few days gave a separation factor\* of 8, which meant that one component was for all intents and purposes pure  $\text{Ne}^{20}$ . On the other hand a several-column apparatus employed by Shrader on  $\text{HCl}$  gave a separation factor of 3 or so after several weeks of running. This great difference illustrates the large effect of the nature of the interaction between the molecules.

It is quite possible to employ several columns in series, the passage of gas from one to the other being achieved by convection. Both glass and metal columns can be used, and the apparatus lends itself to more or less automatic running; no special attention is needed if the water and power supplies are reliable. For this reason it is likely that thermal-diffusion equipment will become a commonplace in most large experimental institutions.

Considerable success has been attained by the method of chemical exchange as developed by Urey and his associates. Although the primary process is apparently quite different from the method of thermal diffusion there is considerable similarity between the two methods in the use of long columns to cascade the primary process. The method of chemical exchange requires considerable research into the equilibrium of various exchange reactions to find one which is suitable for use. By this is meant that, if, for example, we have a gas and a liquid in which heavy and light isotopes are exchanging, an appreciable difference in concentration of light and heavy will exist in the liquid and gaseous phases. To consider a specific reaction:



It is found that when final equilibrium is reached the ratio of the concentration of the light nitrogen isotope as gas to that of the heavy is 1.02, a factor which is appreciable. Now if there can be devised a method of cascading this separation it will not be excessively difficult to obtain from this rather small factor a final very large separation. The method of cascading the process is the familiar method of fractionation. A stream of gas rises up a column to meet a flow of ammonium sulphate down, and in this way there is a continuous tendency for the light isotope to move upward with the gas and the heavy to move downward with the liquid. With enough care and ingenuity very large yields of  $\text{N}^{15}$  of greater than 50 per cent abundance have been secured, and as nitrogen does not exist in a suitable radioactive form for tracer work this makes feasible experimental work which

\* The separation factor is the ratio of ratios of the two isotopes. If  $N_A/N_B$  is the normal ratio of  $A$  to  $B$  and  $S_A/S_B$  the separated ratio, the separation factor is  $\frac{S_A/S_B}{N_A/N_B}$ .



otherwise would be impossible. The method has also been used for oxygen and carbon with good yields, and there is evidence that sulphur can be separated in this way.

During the war revolutionary improvements were made in two methods of isotope separation. The first is the electromagnetic method, which is in essence a huge mass spectrograph designed specially for the purpose of collecting quantity lots of separated isotopes. The products of a part of the plant built for the separation of  $U^{235}$  are now made available for research use, and as a result separated isotopes of boron, silicon, potassium, and others can be obtained from the Isotopes Division of the Atomic Energy Commission. The second method is the gaseous diffusion method. This is in principle much simpler than the thermal diffusion method since it depends on the fact that the molecular velocity at any given temperature depends inversely on the square root of the molecular weight so that the rate of diffusion through a barrier of small holes also depends on the molecular weight. Thermal diffusion is simpler as a laboratory method, first, because of the difficulty of making the barrier (which must have only small holes, comparable in size to the distance between collisions), and second, because of the fact that to make a cascade process it is necessary to pump the gas, whereas in thermal diffusion convection does the cascading. On a large enough scale simple diffusion is successful, but it will almost certainly only be applied to the development of atomic fuels, because only for that purpose is it economically feasible.

One or two "freaks" are also of interest. Deuterium, in which the ratio of masses is so large and the difference of properties so great that, for example, separation by electrolysis is quite easy, is familiar. The other is  $He^3$ , which could readily be separated from  $He^4$  by using the extraordinary properties of ordinary helium at very low temperatures. Ordinary helium, when cooled below 2.5 degrees absolute, becomes a superfluid with very abnormal properties—virtually zero viscosity—and almost any process requiring the flow of helium as a liquid would separate  $He^4$  from  $He^3$ . The reason for this is the symmetry of ordinary helium which requires that it obey the Einstein-Bose statistics, whereas  $He^3$ , with two protons and a neutron, could not do so.  $He^3$  would thus have the normal viscosity of a liquid, and the separation of the two isotopes should be a simple matter.

### Biological use of stable isotopes as tracers

One of the simplest uses of stable isotopes is concerned with the exploitation of simple mixtures. If we have a mixture of a number of



rather similar substances (Rittenberg quotes alpha amino acids, but any mixture which is difficult of separation can be treated in the same way) and desire to know the proportion of one of the constituents present, we may do so without actual analysis as follows. Prepare a small sample of mass  $w$  of one of the amino acids containing a percentage *excess* of, say,  $C^{13}$ , of  $P_{13}$ . This means that if there are in this sample  $N_{13}$  *extra* molecules with the heavy isotope, and  $N_{12}$  ordinary molecules, the value of  $P_{13}$  is given by

$$\frac{P_{13}}{100} = \frac{N_{13}}{N_{13} + N_{12}}$$

Now if the sample is allowed to mix in with the conglomerate and some of the original amino acid is isolated and tested in the mass spectrograph a new value for the percentage excess  $P'_{13}$  will be found. If  $N'_{12}$  is the number of molecules of this particular amino acid present in the conglomerate we can see that

$$\frac{P'_{13}}{100} = \frac{N_{13}}{N_{13} + N_{12} + N'_{12}}$$

and on dividing the two equations by each other we get

$$\frac{P_{13}}{P'_{13}} = \frac{N_{13} + N_{12} + N'_{12}}{N_{13} + N_{12}}$$

If  $W$  is the mass of the amino acid present in the conglomerate, the right-hand side of the above equation is simply  $(W + w)/w$ , and so we deduce that  $W = w(P_{13}/P'_{13} - 1)$ . This means that only a small amount of the amino acid under test need be isolated, and as about 20 mg is ample this is not usually difficult. Notice that no exchange must take place, and that  $P$  and  $P'$  refer to percentage *excesses*.

Rittenberg has used this method to examine the extent of racemization of glutamic acid in tissue. A *d-l* mixture containing excess  $N^{15}$  was added, and the two were separated in samples sufficient to permit the determination of the new excess. In this way the amount of either kind of glutamic acid was determined and the racemization shown to be small.

The use of deuterium, carbon, and nitrogen tracers has necessitated the synthesis of organic compounds of the kind occurring in biological organisms. This difficult task has been attacked, notably by Schoenheimer. Schoenheimer has made an extensive series of syntheses of compounds containing deuterium and nitrogen. The deuterium is



always used in compounds in which it is bound to carbon, and it therefore acts as a tracer for the carbon chain. Nitrogen is used in the form of amino acids. The procedure is first to introduce a small amount of the substance to be studied into an animal, which of necessity is small; after a short time of exposure the animal is killed and a large number of pure compounds isolated from the animal. The presence of excess of deuterium in any of these samples is determined by measurement of density, and by measurement of nitrogen by mass-spectrograph analysis. The fact that different compounds are found to contain the excess isotope indicates the various chemical changes which have taken place.

Schoenheimer, by introducing fatty acids with excess deuterium content, has shown that although the proportion of various fatty acids in the body remains the same there is actually a continuous interconversion of one into the other. A simultaneous synthesis and breaking-down process was observed in which a series of complex fatty acids was built up from simple compounds while complicated substances were broken down into simple ones. The processes are quite rapid. The same feature of complicated interchange, with some means regulating the actual amounts of each substance, was found among the amino acids of the proteins in living animals. From the fate of  $N^{15}$  administered as various amino acids it has been inferred that peptide linkages are continuously opening and closing, and that amino groups can be detached from amino acids while nitrogen can be attached to nitrogen-free substances. The interchange takes place among virtually all the proteins of the animal. A detailed account of Schoenheimer's work is not to be attempted by the authors of this book, as the erroneous description of intricate organic reactions would not add to its value. The fundamental processes taking place in organisms seem to be coming within our comprehension, however, as a result of this type of work.

As the sensitivity and precision of the mass spectrometer are increased it is possible to exploit it for remarkable geological and biological studies. Thus consider the abundance of the potassium isotopes in a rock. The proportion of  $K^{40}$  to  $Ca^{40}$  together with the knowledge of the half-life of  $K^{40}$  (not too good as yet) make it possible to determine the age of the rock. (Such work, which exploits very accurate mass spectrographic analysis with a knowledge of radioactive processes, looks very promising. A pioneer in this field has been Harrison Brown.)

There is not much difference in principle between experiments in which stable isotopes are used and those using radioelements. The two important elements oxygen and nitrogen do not occur in suitable forms



as radioelements, and so they are available only as stable isotopes. In general, if a radioactive isotope is conveniently available, it should be preferred, as the means of detection are so simple. Radioactive isotopes can also be diluted many more times than stable isotopes and so have a rather wider range of application. It is greatly to be desired, however, that several different methods of approach be available to the same problems, for the confirmation one gives to the other is the most certain way to guarantee good experimentation.

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## 10. Nuclear Fission ✓

The story of nuclear fission really begins at the University of Rome in 1934. The neutron had just been discovered by Chadwick in England, and an Italian physicist, Enrico Fermi, with characteristic scientific curiosity, exposed practically all the elements in the periodic table to these uncharged particles. His efforts were rewarded by the discovery of a large number of new radioactive substances. The usual process involved the capture of a neutron by the nucleus in question. This, in turn, often produced an unstable nucleus possessing too much mass for its charge. The atom then proceeded to return to a stable state by emitting a beta ray. This, of course, resulted in an element one unit higher in atomic number than the parent atom. What, reasoned Fermi, would happen if uranium were exposed to neutrons? Recall that in 1934 uranium stood last in the periodic table. If the above mechanism were followed, one should finish with an element of atomic number 93. But in all nature no such element was known to exist. This was an experiment to excite the imagination of a future Nobel laureate. So Fermi and his associates did just that. After an extended exposure to the neutron source, the uranium showed an activity that could be broken down into four half-lives: 10 seconds, 40 seconds, 13 minutes, and 90 minutes, with some indication of still longer ones.

Now there are but three naturally occurring isotopes of uranium, so the appearance of four and possibly more half-lives showed that some unusual process was operating. It was reasonable to guess that one of these activities corresponded to an active form of element 93. Since the idea of a new rare earth series starting with actinium had not yet been conceived, element 93 would appear in the periodic table in the same column with Mn, Ma, etc., and presumably have similar chemical properties. Hence, to test this point, a manganese salt was added to a uranium salt solution which had been irradiated by neutrons. The Mn was then precipitated as  $\text{MnO}_2$ . About one-sixth of the 13-minute



and 90-minute period activity was brought down by this procedure. None of the natural uranium activity appeared. This proved that the active bodies were not chemically like uranium or its immediate daughter products, inevitably present in uranium. It was found further that if small amounts of an isotope of radium or actinium were added to the uranium solution neither would be precipitated with  $\text{MnO}_2$ . This, together with certain other facts, permitted Fermi to conclude that the precipitated activities were not to be associated with any element between atomic numbers 86 and 92, inclusive.

Considering the known nuclear reactions, this was evidence, beyond a reasonable doubt, for believing that at last man had pushed beyond the bounds of nature into the realm of the transuranic elements. Naturally this announcement created a great deal of interest among physicists and chemists, and several became active in further work on the problem. By the end of 1935 it had been pretty definitely established that the bodies responsible for the two longer periods could not be isotopic with any element from mercury to uranium.

Having convinced themselves that these activities had to belong to transuranic elements, three German scientists, Hahn, Meitner, and Strassmann, tackled the job of identifying the elements responsible. After an extensive series of experiments with various times of irradiation, the use of fast and slow neutrons, and numerous chemical tests, they finally proposed a scheme whereby three active uranium isotopes were formed, each of which decayed into successive transuranic elements. By this time, through more careful experiments, the original four half-lives had increased to nine, and elements of atomic number up to 97 had to be called into service to take care of all the findings.

This scheme, though it did seem to cover the experimental results, raised several questions almost as perplexing as the original one of identification. The radioactive yields for each mode of decay required that the prolific isotope  $\text{U}^{238}$  be responsible for all three active uraniums. How was one to explain the formation of three isomeric uranium nuclei of mass 239, two produced by either thermal or fast neutrons, the other by resonance capture of neutrons having energies of a few volts? How could one justify their subsequent decay through the same transuranic elements, but with greatly differing half-lives at each stage? And why should the addition of one neutron to the practically stable  $\text{U}^{238}$  set off a chain firecracker that had to belch up five beta rays before returning to normalcy? It is doubtful that anyone, including the experimenters themselves, relished this solution of the mystery, but no one was able to offer a better answer.



To attempt an explanation of these difficulties, several workers stuck doggedly to the problem. In 1938 two of them, Curie and Savitch, struck a promising clue but unfortunately failed to follow it up. They discovered a new active body in irradiated uranium, this one with a 3.5-hour half-life. It was precipitated with lanthanum as a carrier, thus suggesting that it might be actinium, since lanthanum and actinium appear in the same column of the periodic table. They actually added some actinium to a solution containing the 3.5-hour activity and by fractional precipitation succeeded in changing the relative activity of the two. We now know that the 3.5-hour period is in reality due to an isotope of lanthanum, and the above result might well have been the key allowing Curie and Savitch to be the discoverers of fission. But fate ruled otherwise, and after a further series of experiments they decided that the active substance differed slightly from lanthanum. This left only the already overpopulated transuranic territory in which to dump the 3.5-hour stepchild. In doing so they stressed the difficulty of finding a place for an element having chemical properties like lanthanum in the region of the periodic table just beyond uranium. This added another complication to the already impossible maze.

About this time Hahn and Strassmann re-entered the picture. They repeated the experiments of Curie and Savitch and then went on to discover that, with barium as a carrier, certain activities could be precipitated from an exposed uranium solution which grew into other active elements precipitable with lanthanum. The precipitate they labeled as due to an isomeric radium isotope and the growing activity as due to isomeric actinium nuclei. But, again, difficulties confronted them. To get from uranium to radium, the nucleus involved must lose four positive charges. The most logical way in which this can happen would be by its emitting two alpha particles. These were searched for, but without success. Again the picture darkened, but, as we shall see, it proved to be the darkness before the dawn.

Realizing that it would be difficult to convince responsible physicists that a  $U(n, 2\alpha)Ra$  reaction could take place with slow neutrons, Hahn and Strassmann carried out an elaborate series of tests to prove rigorously that the active atoms in question were truly like radium. Their researches finally reached the point where they could say definitely that radium and barium were the only elements to which the activities in question could possibly belong. All that remained now was to eliminate barium and they would prove their point. To do this they took some of the separated "radium" obtained from irradiated uranium



and added a little barium and  $\text{MsTh}_1$  (an isotope of radium). They now performed fractional precipitations and crystallizations of the kind used for separating radium from barium. They expected that the "radium" atoms would go along with the  $\text{MsTh}_1$  and concentrate in the usual way, thus eliminating barium from consideration. But the expected did not happen. True, the  $\text{MsTh}_1$  concentrated as it should have done, but the "radium" remained uniformly distributed among successive samples. The conclusion, though dumbfounding, was inescapable. The "radium" atoms were really barium. A new concept was necessary. For some reason, when a neutron is added to a uranium atom the union can result in a splitting off of a barium atom. Instead of pushing off into the uncharted transuranic seas, workers had for years been blindly paddling in well-known waters, half-way down the periodic table. It was a discovery to thrill the most sophisticated person and yet at the same time fill even the egotist with a sense of humility. So, it was probably with mixed emotions that Hahn and Strassmann announced their discovery.

If these so-called radium atoms were truly barium, then the immediate daughter activities should be in reality lanthanum. As a double check on their momentous discovery, they proved that this was so.

Why do we attach such great importance to this single discovery? One could dramatically answer "Hiroshima," but from a scientific viewpoint it will be more profitable to consider the packing fraction curve, Fig. 1. Note that the value for stable elements with  $A \approx 118$  is roughly  $-0.0005$ , whereas for uranium the value is about  $+0.0005$ . On this basis, cleaving a uranium nucleus in half to form two lighter nuclei would result in the eventual disappearance of  $[0.0005 - (-0.0005)] \times 236 = 0.236$  unit of mass. According to Einstein's mass-energy equivalence concept this disappearing mass should appear in the form of energy. Relatively speaking this energy release would be tremendous, amounting to more than 200 Mev. The greater part of the release would be imparted to the fragments as translational kinetic energy. Another fraction would appear later as beta and gamma radiation during the return of the fragment atoms, overburdened with neutrons, to a stable form. The magnitude of this process is astounding when it is recalled that the explosion of a TNT molecule releases only a few electron volts, not million electron volts, of energy.

The announcement by Hahn and Strassmann caused a beehive of activity in many places. Nuclear physicists realized that, if such a splitting of the uranium atom took place under neutron bombardment, the energy release could be easily detected by the simplest kind of



research apparatus, present in every nuclear laboratory. The expected was quickly found. Under neutron bombardment uranium *did* split into two lighter fragments. Hahn and Strassmann were entirely correct. In three different ways this was shown.

The most obvious arrangement included an ionization chamber lined with a uranium-containing material and having the output of its amplifier connected to an oscilloscope. In the absence of a neutron source

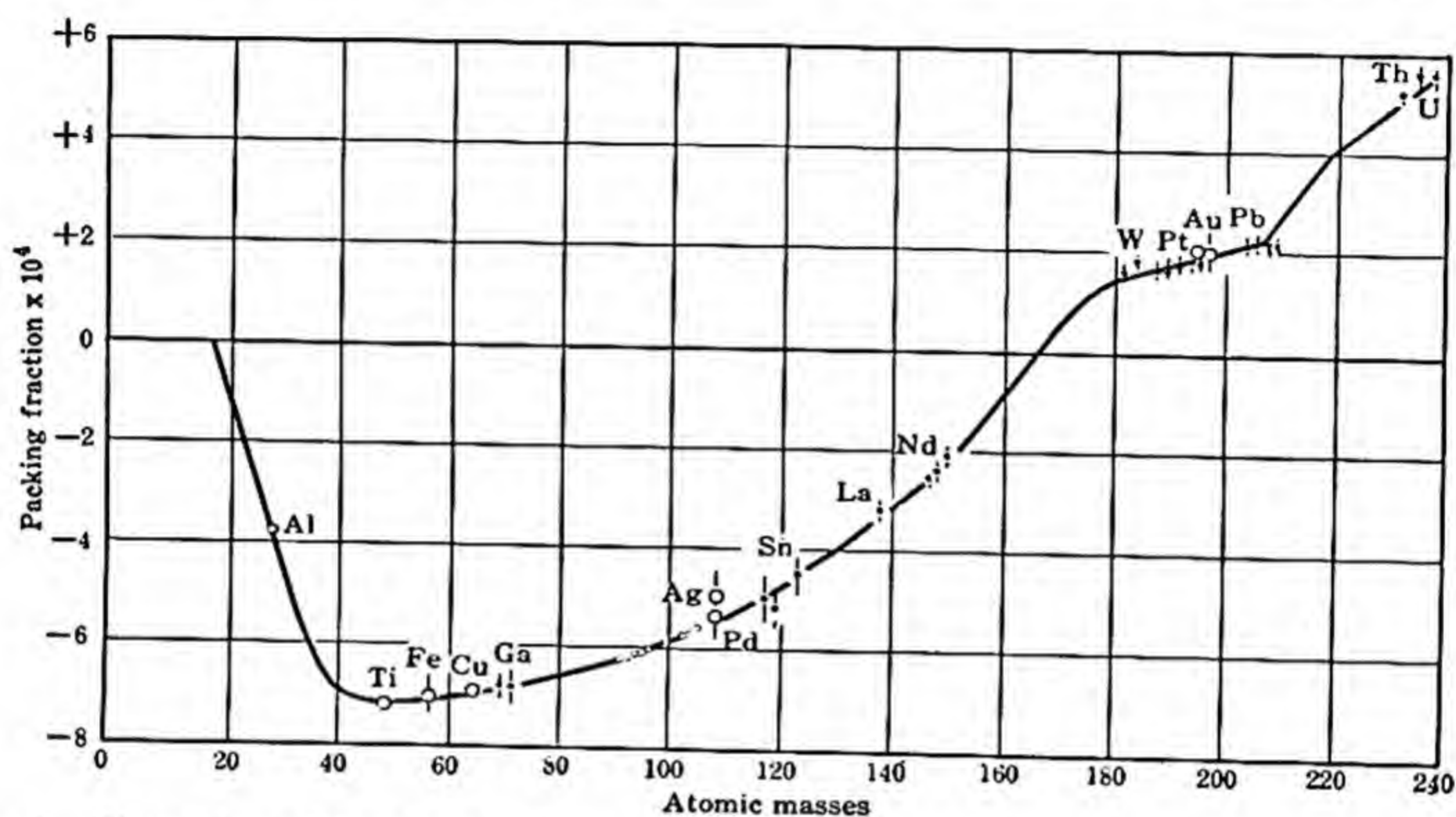


FIG. 1. Plot of packing fraction versus atomic mass from data due to Dempster. The packing fraction is the mass deviation from a whole number divided by the mass. It can be seen to be at a minimum around mass number 50 and high at both very light and very heavy elements.

the pulses on the oscilloscope screen were those caused by the alpha particles from the natural radioactivity of uranium. The energy of these particles is well known, and the size of the pulses could thus serve as a standard of comparison for those due to fission. Whenever the ionization chamber was placed near a neutron source, strange things began to happen on the oscilloscope screen. Huge kicks, judged to represent ionizing particles with energy of 100 Mev or more, appeared. Figure 2 is a representative picture. Indeed, here on display was the greatest man-made nuclear catastrophe yet produced. Stranger than the effect itself is the fact that no one had accidentally stumbled onto the phenomenon years before.

A second picturesque way of demonstrating fission involved use of a Wilson cloud chamber. A thin plate containing a layer of uranium evaporated on mica was fastened inside the chamber. The chamber



was then placed in the neighborhood of a neutron source. Periodic expansions of the chamber were made and photographs taken of each. At first only the thin lines of condensed vapor representing paths of uranium alphas and neutron recoils were seen. Then it happened. Two thick stubby lines of ionization appeared in one picture. It was without doubt the heavy fragments from the splitting of a uranium nucleus. Figure 3 is a reproduction of one such cloud-chamber picture. Final proof that the responsible particle is a very heavy one is seen in the short spurs jutting out from the main tracks. These are

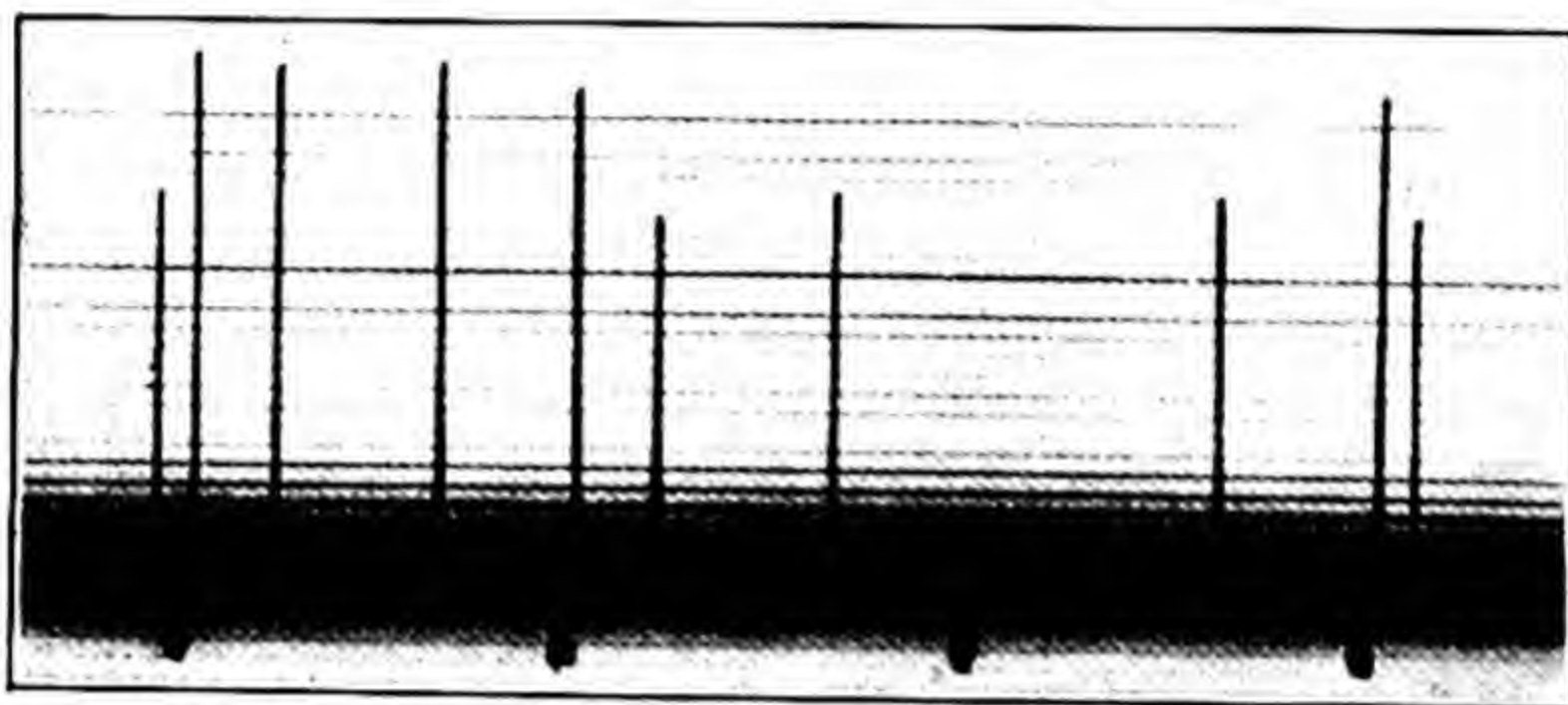


FIG. 2. Photograph, taken by J. R. Dunning, of an oscilloscope screen showing pulses due to ionizing fission fragments.

due to glancing collisions of the fission particle with nuclei of helium, oxygen, or carbon, present in the vapor of the chamber. The proof of the heavy mass of the particle comes from the fact that these collisions do not cause the fragment to deviate noticeably from its straight-line path. In order that the principle of momentum conservation be obeyed, the two parts of the uranium nucleus fly off in opposing directions when it is a slow neutron which touches off the fireworks.

A third way of verifying the explosive nature of the process is less spectacular than the preceding ones but fully as conclusive. Here a layer of uranium is placed close to, but not in contact with, a collector. For the collector Joliot used a cylinder of Bakelite; Meitner and Frisch employed a water surface; McMillan used stacked cigarette papers; Bretscher and Cook used a glass plate. All found what they expected to find; namely, that upon exposure to neutrons the collector became covered with radioactive atoms and that the half-lives agreed with those produced in the uranium itself. The conclusion was unequivocal. These atoms had reached the collector by explosive disruptions of uranium nuclei, for mere capture of neutrons and ejection of beta particles would not have given sufficient energy to heavy atoms



to eject them from the sample. This was direct evidence indicating that the whole group of radioactive "transuranic" elements must be in reality isotopes of an element of lower atomic number. There was one exception to this. McMillan prepared a thin uranium sample from

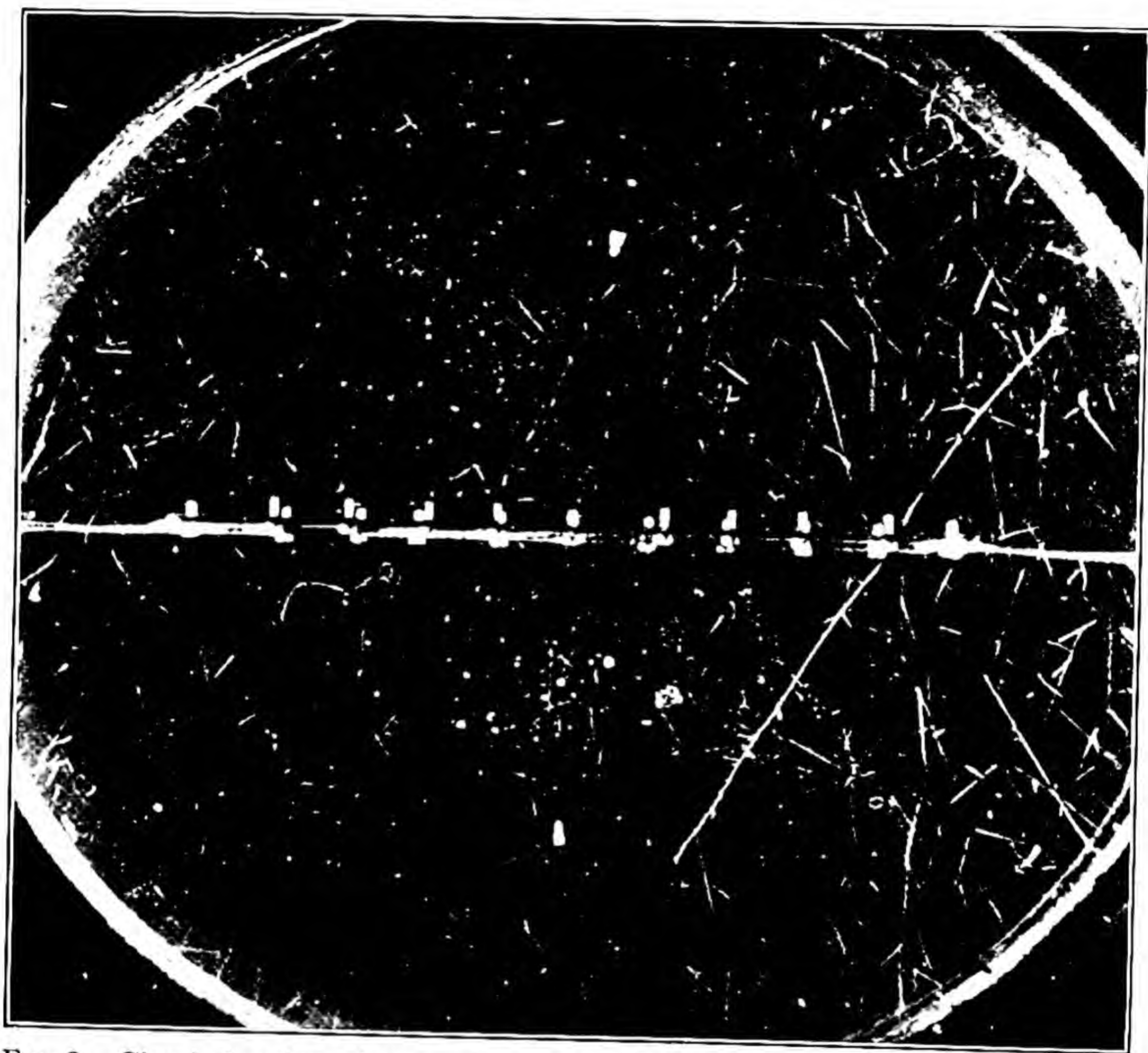


FIG. 3. Cloud-chamber picture of uranium fission showing paths of both fission fragments. Note in particular the spurs branching from the fission tracks. The numerous other tracks in the photograph are the result of fast neutron recoils. (Photograph courtesy of Prof. T. Lauritsen.)

which practically all fission products escape. After exposure to neutrons the uranium layer contained a 23-minute activity. This was attributed to  $U^{239*}$  decaying to a true transuranic element (93), which today is known as neptunium.

These experiments were of an exploratory nature and intended mainly as a proof of the existence of the fission phenomenon. Once it



had been definitely established that uranium did break up, workers settled down to a systematic study of the reasons for and the results of such a process. The discovery of fission cleared up the difficulties encountered earlier in explaining the experimental results. No longer was it necessary to assume  $(n, \alpha)$  or  $(n, 2\alpha)$  reactions for such heavy elements, the formation of  $U^{239*}$  with triple isomery, or the formation of long series of transuranic elements. Fission, however, did raise a multitude of new problems. What isotope or isotopes of uranium were responsible? Did other elements behave similarly? Did the uranium nucleus always break into just two fragments? If so, were they of equal size and charge? Were neutrons released in the process of fission? If so, why was the action not cumulative, i.e., a new neutron causing another fission, and so on, yielding a chain reaction? Could bombarding particles other than neutrons induce fission? All these points will be considered later in this chapter. The most basic question that puzzled scientists, however, was the mechanism whereby a moderate activation of the nucleus could have such startling consequences. The masses of various nuclei show that all massive elements might be broken into lighter fragments with a release of energy. Why, then, are the heavy elements stable in the first place?

### Liquid drop theory of fission

Long before the discovery of fission, different physicists had proposed a liquid drop theory of nuclei. This model proved to be a natural one for explaining the fission process. According to this theory an atomic nucleus can be likened to a liquid drop, having charge  $Ze$  distributed uniformly throughout its volume. The short-range nuclear forces overcome the coulomb repulsion present and cause the droplet to assume a spherical shape as its most stable configuration. These nuclear forces correspond to the cohesive forces acting within a liquid. The total effective energy in a nuclear drop can be represented by

$$\text{Total energy} = -E_n + E_c$$

where  $E_n$  represents the energy due to the nuclear attractive forces, and  $E_c$  is the energy occasioned by the electrostatic repulsion. If the total energy is negative, the drop is stable and the absolute value is referred to as the binding energy. The energy of cohesion  $E_n$  can be broken into two terms:

$$E_n = E_v - E_s$$



$E_v$  being a so-called *volume* energy, and  $E_s$  a *surface* energy.  $E_v$  is taken as proportional to the total number of particles present in agreement with the saturation character of nuclear forces. Clearly this term alone gives an incomplete description of the situation since the particles near the surface, unlike those within the drop, are not attracted equally on all sides. For this reason the quantity  $E_s$  must be subtracted to yield a net value in accord with reality.

The total energy thus assumes the form

$$E_t = -E_v + E_s + E_c$$

The question now arises as to what would happen if the nuclear drop were in some way displaced from its spherical shape.

Bohr and Wheeler, among others, considered this question in some detail. First they examined the effect of a *very small* deformation on the spherical droplet. They agree that to a first approximation  $E_v$  remains unchanged. The electrostatic energy will decrease, but this will be opposed by an enhancement in  $E_s$  due to the increased surface. The net change in potential energy is found to be proportional to:

$$\frac{2}{5} E_s \left( 1 - 0.022 \frac{Z^2}{A} \right)$$

where  $E_s$  is the surface energy of the undisturbed drop,  $Z$  is the atomic number, and  $A$  is the mass number.

As long as the quantity within parentheses is positive the result will be an increase in total energy (decrease in binding energy). This implies that once the distorting force is removed the droplet will return to its original shape. But obviously, for a nucleus having  $Z^2/A > 45.4$ , this restoring tendency is not present, and under the slightest deformation the nucleus will proceed to divide. By an approach involving the experimental fission threshold for  $U^{239*}$  ( $U^{238} + \text{neutron}$ ) Bohr and Wheeler deduced a more accurate value for  $Z^2/A$  to be 47.8. Now  $Z^2/A$  for uranium is around 36.0 which is well below the limit. This tells us that a uranium nucleus, and hence all lower nuclei, should be stable against very small deformations.

But what is the situation if we consider the possibility of *relatively large* deformations? An analysis of the problem reveals that, although at first the surface energy of the drop increases more rapidly than the electrostatic energy decreases, this situation becomes reversed as the drop assumes more and more an ellipsoidal shape. In fact beyond a certain deformation the total energy actually begins to decline. The



point at which the total energy reaches a maximum and starts to decrease is of extreme importance, since a nucleus once subjected to this deformation will then proceed to fission without further assistance. The actual behavior is as pictured in Fig. 4. The mass of  $U^{236}$  is smaller by 1.913 mass units than that of 92 protons + 144 neutrons.

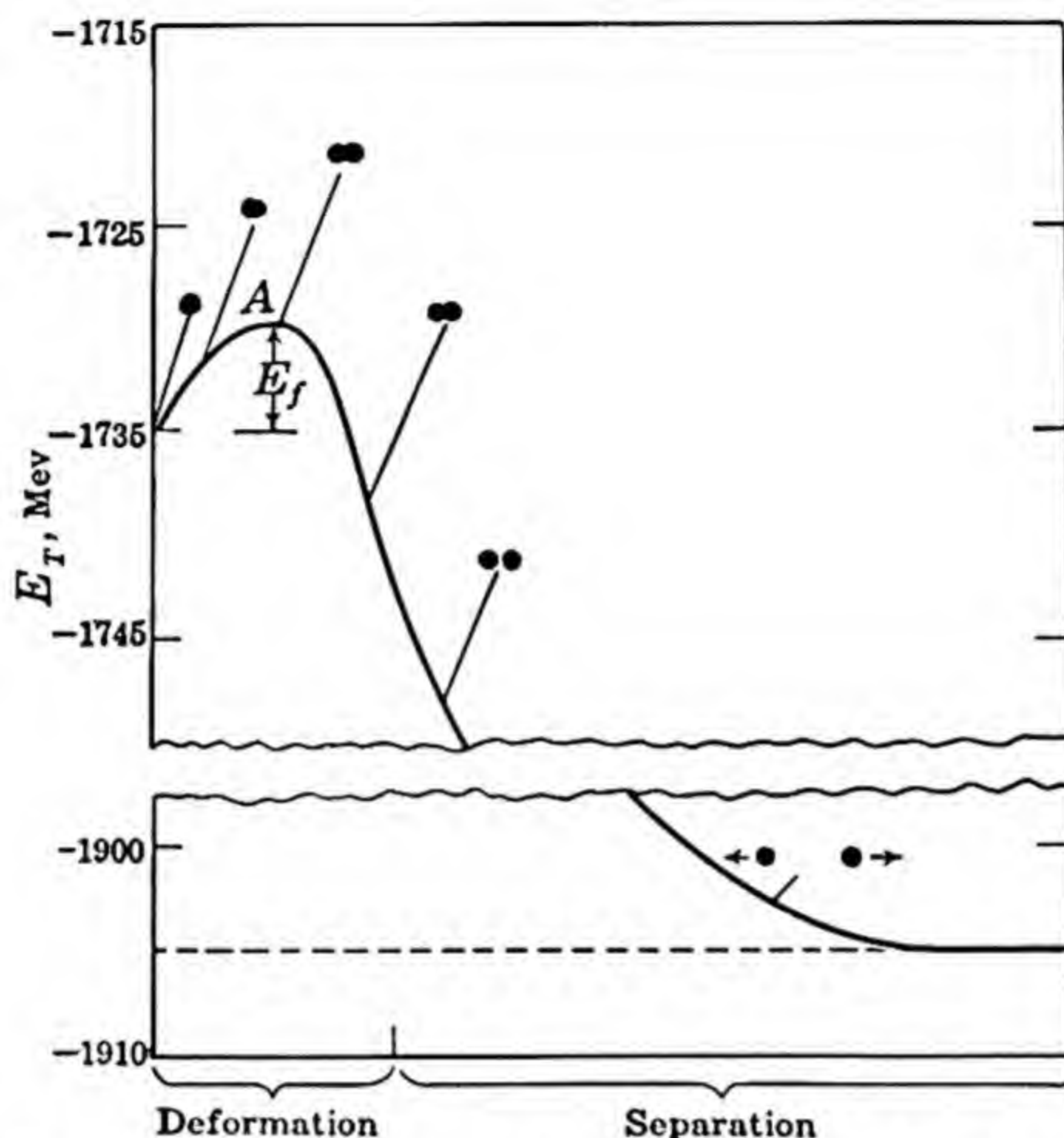


FIG. 4. A plot illustrating the stages through which a  $U^{236*}$  nucleus progresses in the act of fission. Though stable against small deformation, the nucleus once taken to the critical point *A* will proceed to divide without further assistance. Activation energy  $E_f$  (5.2 Mev) must be supplied to produce this critical deformation. The resulting fragments will possess much less total potential energy than the original  $U^{236*}$  nucleus. This difference, amounting to around 170 Mev, goes mainly into kinetic motion of the fragments themselves. In returning to a stable condition by  $\beta$  decay each fission pair eventually releases another 20 Mev, making a total of some 190 Mev for the complete energy release.

Thus we say it has a total energy of  $-1735$  Mev or a binding energy of 1735 Mev. The nucleus is in a stable configuration as long as the energy of deformation remains less than  $E_f$ . But once this value is equaled or surpassed the nucleus rides over the potential barrier and coasts down the other side, splitting into two fission fragments. The binding energies of the product nuclei on the average will total about 1905 Mev, giving an energy release  $\Delta E$  of roughly 170 Mev for the primary fission process (K.E. of fragments, 160 Mev; K.E. of 2 secondary neutrons, 5 Mev;  $\gamma$  radiation, 5 Mev).



If the theory is to yield quantitative information there is obviously the need for a method of evaluating the activation energy  $E_f$  for various nuclei. Because of the mathematical complexities in dealing with large deformations of a sphere, a general solution of this problem is practically impossible. However, the problem becomes relatively simple in two special cases (1) for  $Z^2/A$  near  $(Z^2/A)_{\text{limiting}}$ , and (2)

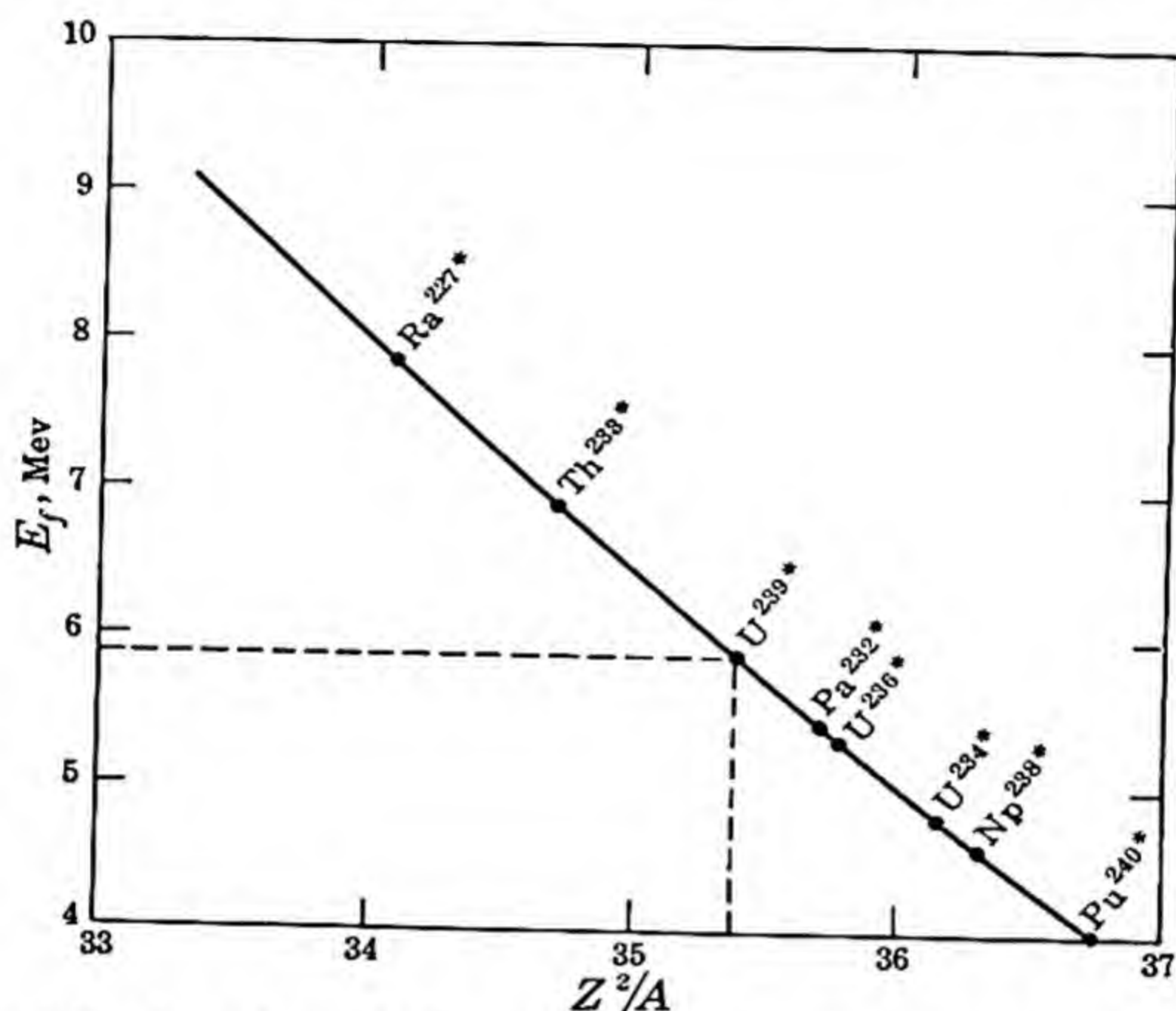


FIG. 5. Bohr-Wheeler plot of  $E_f$  versus  $Z^2/A$  for the region of heavy nuclei. The curve has been fitted to agree with the experimental value of  $E_f$  for compound nucleus  $U^{239*}$ . In all cases the designation refers to the compound nucleus, i.e., target nucleus plus a neutron.

for  $Z^2/A$  near 0, i.e., practically no charge on the sphere. Bohr and Wheeler determined  $E_f$  versus  $Z^2/A$  for  $Z^2/A$  near zero and also for  $45.4 < Z^2/A < 47.8$ . They extended these plots so that they met to give a smooth fit, thus establishing the curve over the region inaccessible to easy calculation. Figure 5 shows a section of the Bohr-Wheeler curve covering the region of greatest interest. The curve has in effect been fitted to agree with the experimental value of  $E_f$  for  $U^{239*}$  ( $U^{238} + \text{neutron}$ ). By means of the curve it is possible to estimate  $E_f$  for other heavy nuclei. Let us now compare some of these values with experiment.

As we have seen, fission is normally induced through the addition of a neutron to a suitable nucleus. This supplies an amount of energy equal to the binding energy of a neutron in the (compound) nucleus



plus the kinetic energy of the incoming neutron. For thermal neutrons the latter term may be neglected. In general, various modes will compete for the excitation energy  $E_a$  acquired by the compound nucleus. This energy may (1) be reconcentrated on a single neutron; (2) be radiated away as a photon; or (3) go into energy of deformation. Processes (1) and (2) will result in neutron and gamma-ray emission, respectively. If  $E_a \gtrsim E_f$ , process (3) can give rise to fission.

Now it is a fact that the energy of binding for a neutron in any heavy nucleus has a value close to 5.4 Mev (odd number of neutrons in nucleus) or 6.4 Mev (even number of neutrons in nucleus).

With this criterion and the Bohr-Wheeler plot, the following table has been compiled to compare the excitation energy  $E_a$  supplied to a nucleus by capture of a thermal neutron with  $E_f$  for the same nucleus. Whenever  $E_a \gtrsim E_f$ , thermal neutron fission may be predicted. This table lists most of the isotopes near the heavy end of the periodic table (both natural and artificial) which are readily obtainable in macroscopic amounts.

It is gratifying to note that except for  $\text{Np}^{237}$  the theoretical prediction is borne out. However, to put things in proper perspective it must be noted that for certain other artificially produced nuclei the theory disagrees with experiment.

In an effort to improve matters, Present, Reines, and Knipp actually calculated the  $E_f$  versus  $Z^2/A$  curve down to  $Z^2/A = 38.3$ . Metropolis and Frankel, using the ENIAC (electronic numerical integrator and calculator), extended the curve to  $Z^2/A = 31.1$ , well beyond the region of greatest interest. These refined calculations bring some of the anomalies into line, but certain others persist. Nevertheless it must be admitted that the nuclear drop theory has gone far in explaining the process of fission.

One fact that has so far resisted explanation is the asymmetry of fission. Everything points to the fact that the maximum  $\Delta E$  in binary fission should occur for a division into two equal fragments. Since physical systems endeavor to attain a state of minimum potential energy, one might argue, a priori, that this mode of splitting would be most probable. In support of this hypothesis the drop theory predicts that  $E_f$  will be smallest for a symmetrical deformation of the nucleus. Yet we will see presently that in the most common mode of division the fragment masses stand in a ratio of  $\approx 2:3$ , an even split being realized but rarely. However, this may not actually represent a failure of the nuclear drop theory since it gives no clue as to what

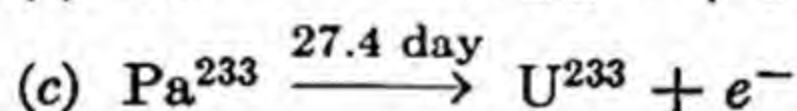
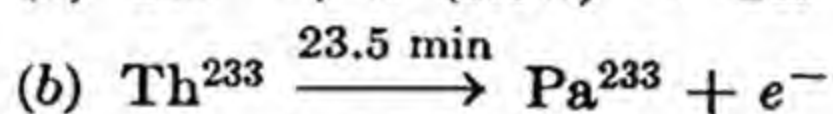
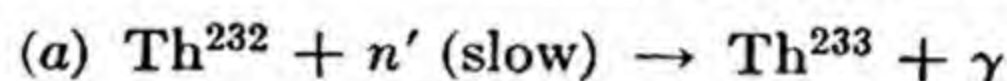


TABLE 1

TARGET NUCLEUS	ORIGIN	COMPOUND NUCLEUS	NUMBER OF NEU- TRONS IN COMPOUND NUCLEUS	$E_a$ (Mev)	$E_f$ (Mev)	FISSIONABILITY WITH $kT$ NEUTRONS	
						PREDICTED	FOUND
$U^{233}$	1	$U^{234}$	142	6.4	4.6	Yes	Yes
$U^{235}$	Occurs in nature	$U^{236}$	144	6.4	5.25	Yes	Yes
$U^{238}$ *	Occurs in nature	$U^{239}$	147	5.4	5.9	No	No
$Th^{232}$	Occurs in nature	$Th^{233}$	143	5.4	6.9	No	No
$Pa^{231}$	Occurs in nature	$Pa^{232}$	141	5.4	5.4	No	No
$Np^{237}$	2	$Np^{238}$	145	5.4	4.6	Yes	No
$Pu^{239}$	3	$Pu^{240}$	146	6.4	4.0	Yes	Yes

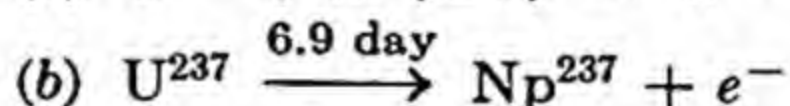
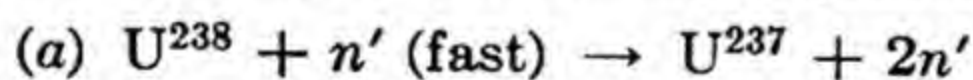
\* Bohr-Wheeler curve fitted at this point.

1. If a mass of thorium is placed in a chain-reacting pile, the following sequence of reactions will occur to produce  $U^{233}$  in considerable quantities:



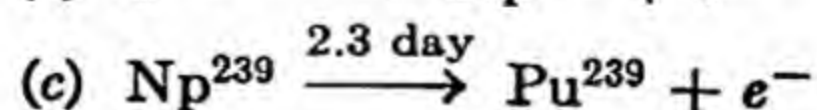
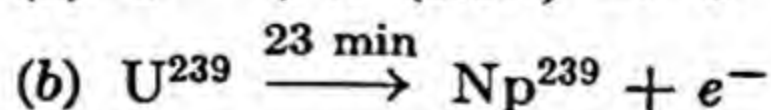
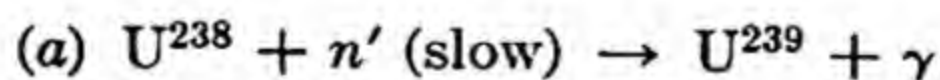
The  $U^{233}$  can then be separated from the Th mass by ordinary chemical processes.

2. In a natural uranium pile there will be a considerable flux of fast neutrons. Some of these will produce  $Np^{237}$  by the following sequence of reactions.



It has been stated that the Hanford piles produce 0.1 per cent as much  $Np^{237}$  as  $Pu^{239}$ .

3. The Hanford piles were built for the purpose of producing this isotope by the following reactions.





will happen beyond the critical point. Perhaps the deformation is symmetrical up to the critical point, and then as the necking down proceeds asymmetry develops prior to the actual division. Recently Frenkel has attempted an explanation on the basis of barrier penetration. This may be a potent argument in the case of spontaneous fission, but for induced fission the excitation energy raises the nucleus above the barrier and hence penetration appears unnecessary. Evidence has accumulated which indicates that closed shells of neutrons and protons occur in certain nuclei, making them particularly stable. Mayer suggests that closed shells of 50 and 82 neutrons may be employed to explain asymmetric fission, the most probable division of the compound nucleus being such that one fragment contains at least 50 neutrons, the other at least 82. Whatever may be the correct explanation it seems evident that this phenomenon of unequal division deserves further attention.

Our discussion of the fission mechanism has already answered some of the questions raised earlier. The following summary of the exhaustive experimental work on the fission process will cover the remainder in some detail.

Before the blackout on publication of fission research occasioned by World War II, the following facts were common knowledge all over the world.

1. Isotope  $U^{235}$ , making up 0.7 per cent of ordinary uranium, is entirely responsible for the thermal neutron fission in uranium.  $\sigma_f(U^{235})$  was estimated as  $\approx 500$  barns. With increasing neutron energy  $\sigma_f$  was found to decrease as  $1/v_{\text{neutron}}$ .

2. Neutrons with energy  $\gtrsim 1$  Mev produce fission in  $U^{238}$  with  $\sigma_f(U^{238}) = 0.5$  barn for neutrons of 2.4 Mev.  $U^{238}$  atoms capture neutrons with energy  $< 1$  Mev to become  $U^{239}$  atoms. (The ultimate fate of  $U^{239}$  was unknown before World War II.) There is a strong resonance [ $\sigma_a(U^{238}) \approx 1200$  barns] for this capture process at  $E_{\text{neutron}} = 5 - 25$  electron volts.

3.  $Th^{232}$  and  $Pa^{231}$  were also found to be fissionable, but only with fast neutrons. None of the lower elements gave positive results even when irradiated with neutrons so energetic that  $E_a$  was well in excess of the Bohr-Wheeler  $E_f$ . [However, with the very energetic neutrons (84 Mev) from the 184-inch California frequency-modulated cyclotron it has been found possible to induce fission in elements as low as tantalum (73), and with energetic protons even copper seems to fission.]

4. Fission could also be produced in U and Th by high-energy alpha particles, deuterons, protons, and gamma rays.



5. The fission process often occurs within  $5 \times 10^{-13}$  second after neutron capture.

6. Secondary neutrons were indeed found to be released as a by-product of the fission process. The majority of determinations gave values for  $\nu$  (number of secondary neutrons per fission) ranging between 2 and 3.

7. It was found that 99 per cent of the secondary neutrons are emitted within 0.001 second after capture of the primary neutron. The remaining 1 per cent are given out with several definite half-lives, ranging from 0.1 second to 45 seconds (postwar limits, 0.05 second to 55 seconds). It was almost certain that these delayed neutrons do not arise from the primary fission process but are emitted by a fission fragment somewhere along its beta-decay chain. However, none of the responsible bodies had been identified prior to World War II. It will be seen in the next chapter that these delayed neutrons, though relatively small in number, play a very important role in the control of thermal piles.

8. There were known to be many possible fission modes. Practically every element between  $_{34}\text{Se}$  and  $_{57}\text{La}$  was represented in the fission products. Essentially the same bodies are found for (a) slow neutron fission of U, (b) fast neutron ( $> 1$  Mev) fission of U, and (c) fast neutron fission of Th, thus emphasizing the statistical nature of the process.

9. Fission normally results in asymmetric division. The fragments fall into a light and a heavy group. The general characteristics of both groups are listed below. These data are for the compound nucleus  $\text{U}^{236*}$ , but the picture would be almost identical for  $\text{U}^{239*}$  or  $\text{Th}^{233*}$ .

TABLE 2

	LIGHT GROUP	HEAVY GROUP
Spread in kinetic energy	75–115 Mev	40–80 Mev
Average kinetic energy	95 Mev	60 Mev
Maximum range	2.2 cm air	1.5 cm air
Most probable $A$ *	91	143

\* This assumes two prompt neutrons per fission and the conservation of momentum. The latter condition requires that  $E_L A_L = E_H A_H$ .

10. It had been found by Flerov and Petrjak that uranium fissions spontaneously but at a very slow rate. Their value of the half-life for this process is  $10^{17}$  years.

It should be emphasized at the outset that, with the exception of the fissioning of elements below thorium by energetic particles, nothing has been published since World War II which would tend to nullify any point in the previous summary. Certain numerical values have been



modified slightly in the light of more precise studies, and progress has been made in extending our general knowledge of this phenomenon. The following summarizes our present knowledge of the fission process.

### ✓ Delayed neutron emitters

In the case of  $U^{235}$  fission the delayed neutrons have now been found to fall into six groups. These groups have the following half-lives: 55 seconds, 22 seconds, 4.5 seconds, 1.6 seconds, 0.4 second, and 0.05 second. The 55-second activity has been assigned to  $Xe^{137}$  and the 22-second activity to  $Kr^{87}$ . The 4.5-second activity is also due to a Kr isotope, but the mass value is not known. The others are as yet unassigned. In explaining the phenomenon for the 55-second period one supposes that when  $I^{137}$  decays to  $Xe^{137}$  by emission of a beta ray, the  $Xe^{137}$  nucleus is left in a state with excitation greater than that corresponding to the binding energy of a neutron. Hence a neutron is immediately ejected, leaving  $Xe^{136}$ . The  $I^{137}$  has a 55-second period, which of course will also be the apparent neutron period. A similar argument holds for  $Kr^{87}$ ,  $Br^{87}$  being the mother atom. Delayed neutrons account for 0.76 per cent of the secondary neutrons from  $U^{235}$  fission.

### Energy distribution of fission fragments

Considerable work was done on this problem during World War II both in this country and in Germany. The results of Fowler and Rosen are typical of those obtained. They used an ionization chamber inside which was mounted a 10-mil platinum foil plated with an extremely thin coating of 94 per cent  $U^{235}$ . The ionization pulses were recorded on a ten-channel discriminator, each channel having such a width as to accept pulses over a range of 0.8 Mev. It was possible to shift the bias of all the channels by the same amount so that the entire energy spectrum could be covered, nine points at a time. Figure 6 shows the results obtained for both slow (thermal) and fast ( $10^3$  to  $10^6$  volts) neutrons. Four main conclusions may be drawn from these plots, most of which merely refine prewar knowledge of the fission process: (1) The most likely fission mode is asymmetrical in character. (2) The energy distribution of the fission products is practically identical for both slow and fast incident neutrons. (3) The most probable kinetic energy of the two fragments per fission is about 155 Mev. (4) The fact that the heavy particle group (low energy) embraces a wider spread in energy provides convincing proof for the statement that at least up to a point the energy release is greater, the more sym-



metrical the splitting. This follows directly if one assumes that momentum is conserved in the fission process.

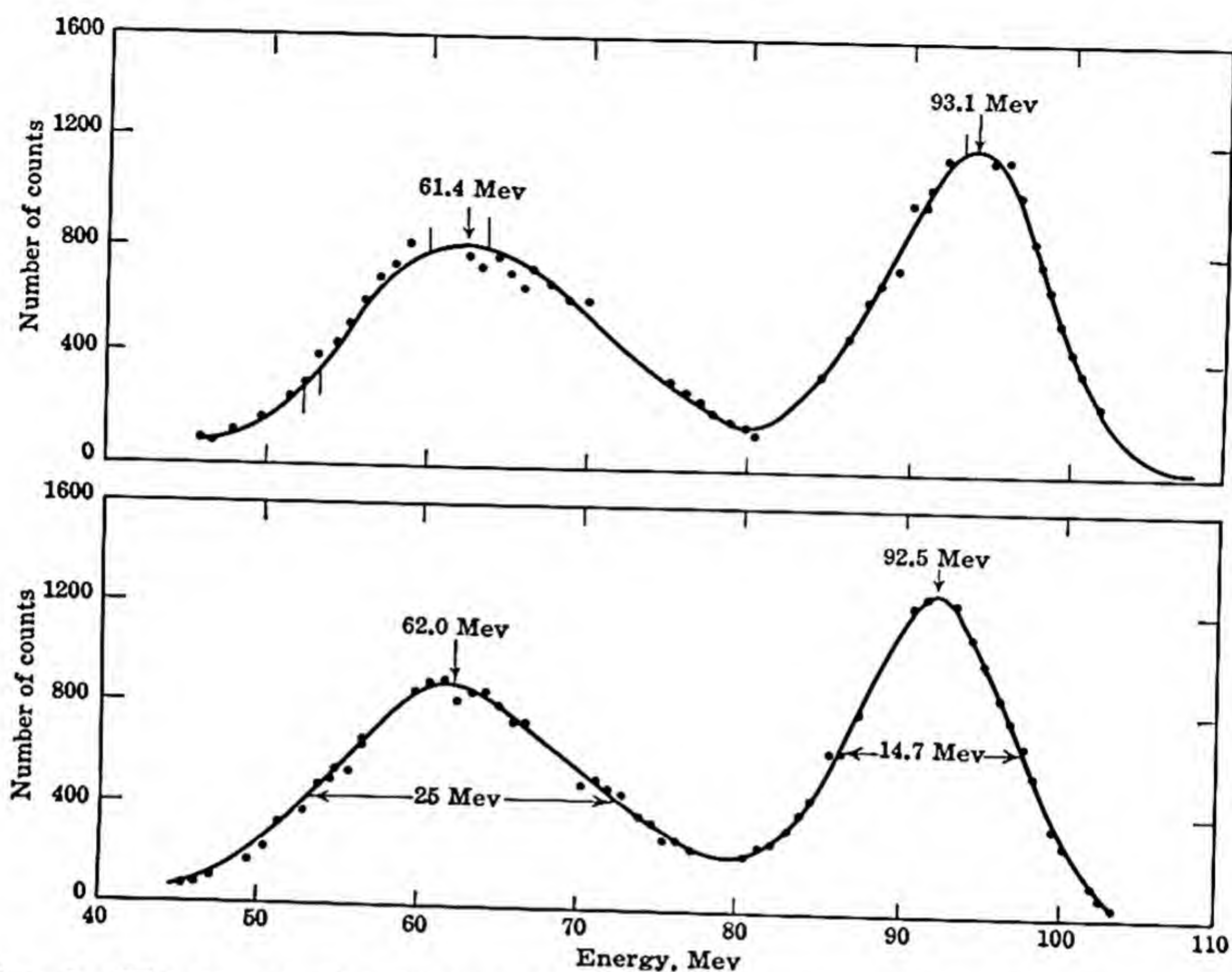


FIG. 6. The energy distribution of fission fragments as determined by Fowler and Rosen. The upper curve is for fast neutrons, the lower for thermal.

### Distribution of mass in fission products

By means of a double ionization chamber whereby the energy of each fragment for a single fission event is measured, one can, assuming momentum conservation, arrive at a curve describing the mass distribution in fission. However, this information may be obtained more accurately, albeit with much more labor, by chemical methods. The procedure here is to take a mass of uranium which has been exposed to neutrons and separate the maze of activities into the component elements. By comparing the decay characteristics with known activities belonging to each particular element, one is usually able to make a unique mass assignment. The mass spectrometer can also be very helpful in this connection. Figure 7 presents the fission yield (the percentage of fissions leading to a particular mass being formed) versus the mass number for  $U^{235}$  (compound nucleus) fission. The



most interesting feature of this plot is the evidence for a slight amount of symmetrical division, i.e., for masses around 117. Energy measurements on the fission pairs failed to indicate this. Since masses in this range apparently are formed but once in every 1500 fissions it is not

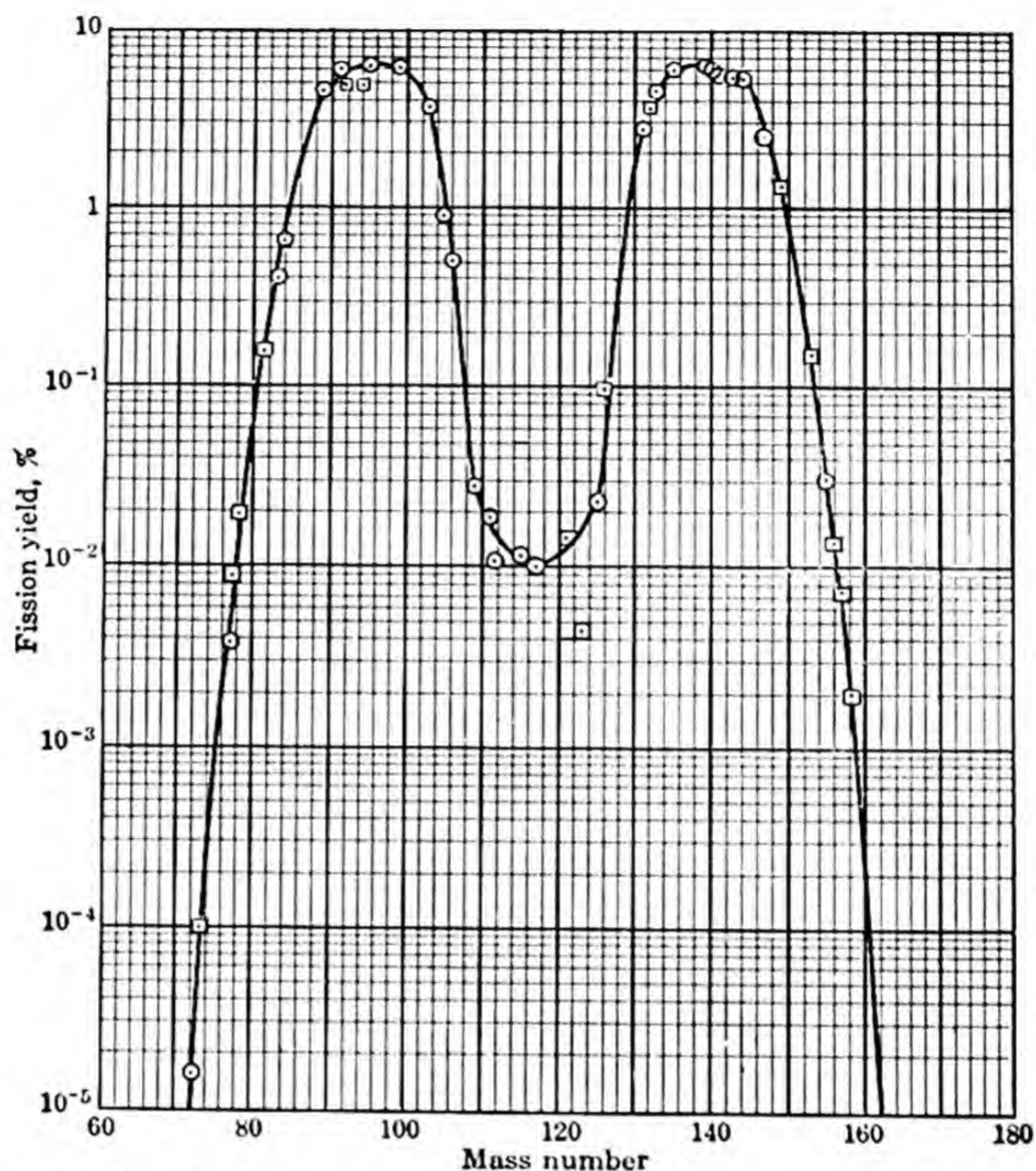


FIG. 7. Yields of  $U^{235}$  fission product chains as a function of mass, published by the Plutonium Project. This curve was established through exhaustive studies on fission products separated by chemical methods into their component activities.

surprising that this phenomenon escaped detection by the ionization chamber technique. With the chemical method it is possible to detect masses formed as rarely as once every  $10^7$  fissions.

### Distribution of charge in fission

The chemical nature and hence atomic number of most fission products has been thoroughly explored. However, in the vast majority of cases this tells us nothing of the initial charge on the fission fragment, since the latter changes atomic number several times as it progresses



down a decay chain toward stability. The fragments are highly excited initially, and as a consequence short half-lives will ordinarily be associated with the first beta decays. This emphasizes the difficulty of securing information on the initial charge distribution.

Four theories have been advanced to explain charge division in fission.

1. The simplest hypothesis is to assume that the charge divides in direct proportion to the fragment masses, i.e., the charge to mass ratio before and after fission is the same.

2. Present has calculated the expected charge distribution on the assumption that in nuclei one expects to find a greater density of protons near the surface than in the interior, owing to their mutual electrostatic repulsion. This assumption leads to the conclusion that the smaller fragment will possess a greater charge than if hypothesis 1 is adopted.

3. Wigner and Way have proposed that the charge distribution at the moment of fission will be such that the total potential energy of the two fragments (electrostatic repulsion plus the subsequent available beta-decay energy) shall be a minimum.

4. Coryell et al. postulate on purely empirical grounds that the fissioning nucleus divides its charge so that the length of the subsequent beta-decay chain shall be roughly the same for all species.

No definite choice can yet be made among these possibilities, but some light has been thrown on the matter by a consideration of the fission yield of shielded isotopes. These are fission bodies possessing a  $Z$  one unit greater than that of a stable isotope of equal mass.  $^{82}_{35}\text{Br}$  is a shielded isotope since  $^{82}_{34}\text{Se}$  is stable. Obviously a measurement on the activity of  $\text{Br}^{82}$  formed in fission will give the *true* fission yield of this body because no other fragment of mass 82 can decay into  $\text{Br}^{82}$ .  $\text{Rb}^{86}$  and  $\text{Cs}^{136}$  are also in this category. The  $\text{U}^{235}$  fission yields of these three isotopes are known, and careful consideration of the data tends to favor hypothesis 1 or 4 although the information is too meager to permit a clear-cut decision.

### Ternary and quaternary fission

Referring again to the packing fraction curve, it is seen that division of a uranium nucleus into three equal parts would actually yield a greater energy release than occurs in binary fission. Fission into four fragments is likewise exoergic (meaning it can occur with release of energy) but less so than for two- or three-particle fission. Calculations indicate that  $E_f$  for ternary fission is not greatly different from



that of binary division. Although fission into three approximately equal fragments seems to occur but once in every 150,000 splittings, several workers have found that about one out of every 250  $\text{U}^{235}$  fissions is accompanied by an energetic alpha particle. For  $\text{Pu}^{239}$  fission the occurrence of an alpha is about half this frequent. Wollan and co-workers find that most of the alphas are emitted at right angles to the main fission fragment paths. This indicates that the alpha is released from the necked-down part of the dividing nucleus as a part of the primary process. Some evidence has been found for quaternary fission occurring about once out of every 3300 fission events.

### Range of fission fragments

Figure 8 depicts the range in centimeters of air equivalent for the fragments from  $\text{Pu}^{239}$  fission as a function of mass. These data were

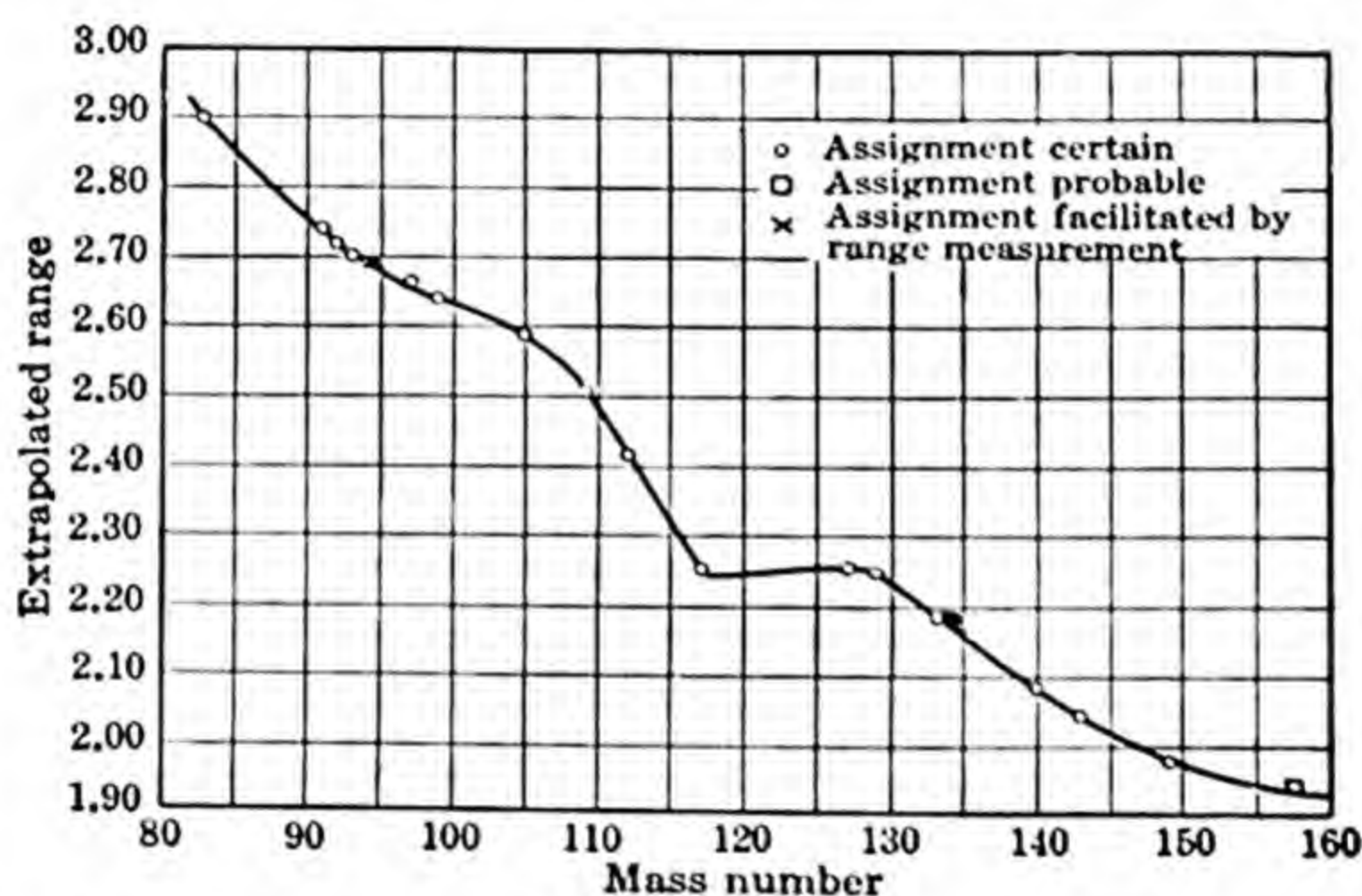


FIG. 8. Extrapolated range of  $\text{Pu}^{239}$  fission fragments (centimeters of air) as a function of mass number. These data were obtained by a clever multiple foil technique (see text). One can combine these data with other information on the total energy release from Pu fission to confirm Bohr's prediction that  $R \propto E^{1/2}M^{-1/6}$ .  $R$  = range of fission fragment of mass  $M$  having energy  $E$ . The data are due to Kateoff, Miskel, and Stanley.

obtained by Kateoff and co-workers by identifying and counting the activity of fission fragments caught on a series of Zapon films placed progressive distances away from a thin Pu layer bombarded with neutrons. Kateoff interprets the humps in the curve at  $A \approx 107$  and 128 as indicating that the greatest energy release occurs for a splitting into fragments having a mass ratio near 1.2. It might be noted that owing to their highly ionized nature fission fragments interact with matter through which they pass in a manner quite different from that exhibited by protons or alpha particles.



### Origin of prompt fission neutrons

By a measurement of neutron coincidences about a fission source as a function of the angle between two neutron counters it has been shown that on the assumption of two neutrons per fission the likelihood is greater that each fragment contributes one neutron than that both should arise from the same fragment.

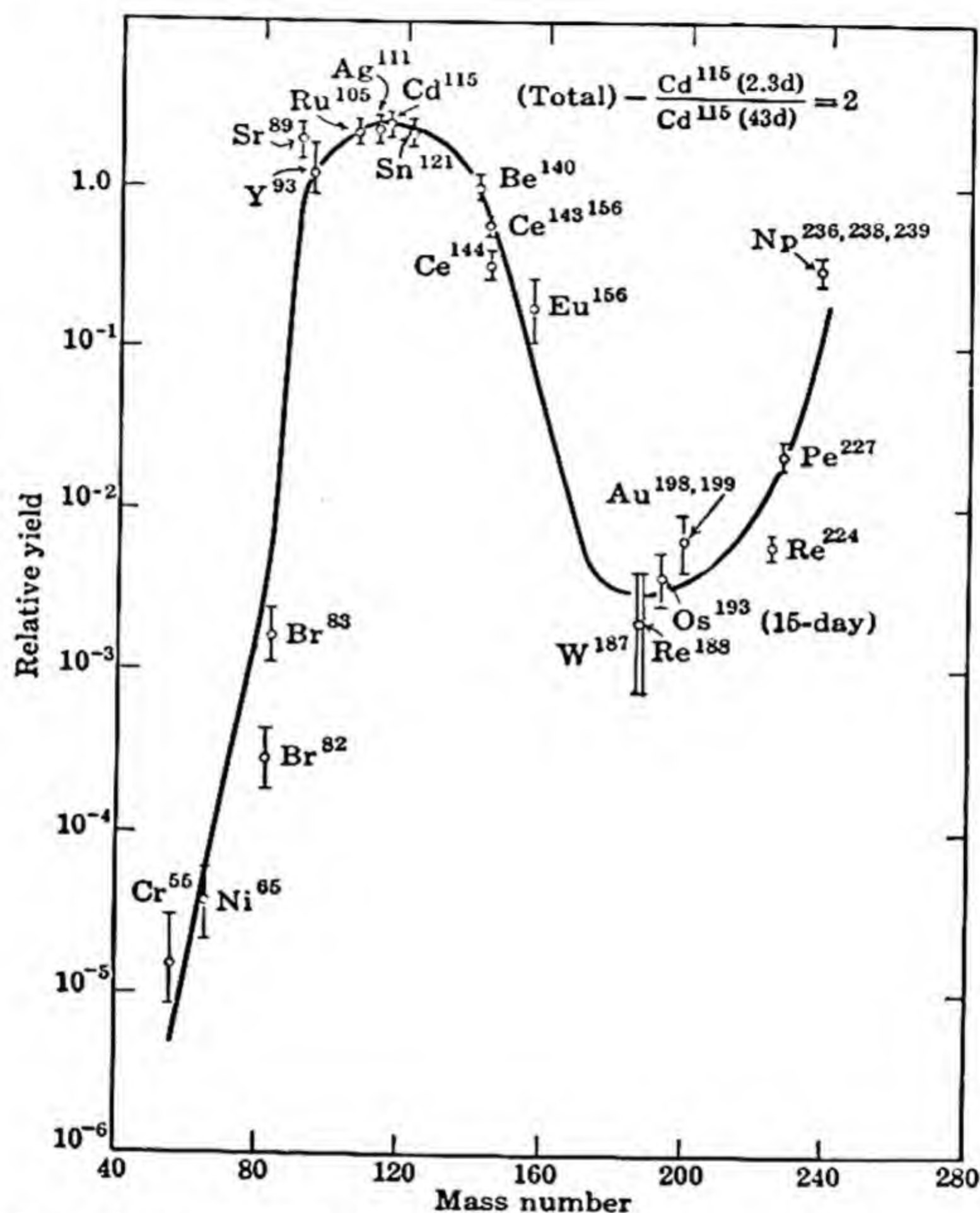


FIG. 9. Relative yield curve obtained by O'Connor and Seaborg for products from 380-Mev helium ion bombardment of uranium. Contrast this curve with Fig. 7.

### Fission induced by extremely energetic particles

Once the 184-inch California frequency-modulated cyclotron had been put in operation some very striking results were obtained concerning fission induced by energetic particles. These studies employed 380-Mev alpha particles, 190-Mev deuterons, and 84-Mev neutrons. Figure 9 plots the activities detected in a mass of ordinary uranium after bombardment with 380-Mev alphas. This curve, in sharp con-



trast to Fig. 7, shows but a single fission peak and a continuous yield of radioactive products for the entire range of elements from Np down to  $Z \approx 25$ . It is thought that the products from  $A \approx 239$  down to  $A \approx 180$  are produced by spallation (a splintering off of small nuclear fragments), whereas those below 180 are true fission products. The single fission peak found here was not too surprising since earlier work had shown that as the energy of the bombarding particle increased the valley between the two mass peaks became more and more shallow. It is proposed that in the present instance the highly excited compound nucleus normally fissions into two equal fragments before the nuclear particles have an opportunity to rearrange themselves into a more energetically favorable configuration, thus retaining in each particle the neutron/proton ratio of the parent nucleus. The fact that the peak in the fission curve comes at a point several mass units below one-half the original mass implies that several (maybe a dozen) neutrons may be boiled off before fission occurs. A similar interpretation has been given to the single peak mass curve from the fission of Bi under the influence of 190-Mev deuterons or 84-Mev neutrons. Interestingly enough the evaporation of a dozen neutrons from a Bi nucleus leaves the residual body with a  $Z^2/A$  approximately equal to that for  $U^{235}$ , so according to the Bohr-Wheeler concept only a very moderate residual excitation is then demanded to effect fission. It is clear, however, that no practical release of energy can be achieved by the fission of Bi and similar nuclei since the emitted particles do not possess sufficient energy to produce further fission in such elements.

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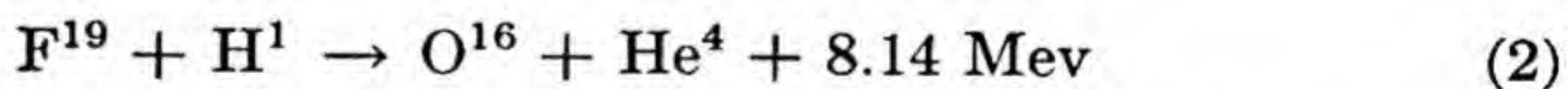
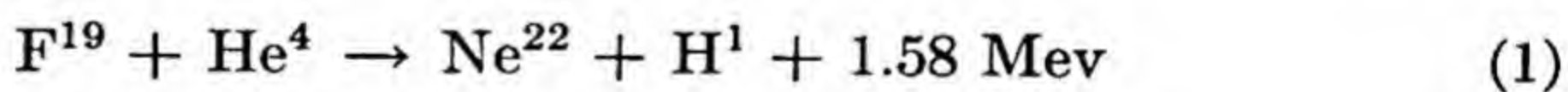


## 11 · Nuclear Chain Reactions

Through the agency of artificial disintegration man has been able to release nuclear energy since 1919. How then does one explain the lapse of twenty-three years before he put this force to work on a practical scale? The answer is simple. Heretofore all transmutation processes were isolated events and the resulting products were simply not able to continue the process. In other words, it was not possible to bring about a nuclear chain reaction.

Chain reactions are not new. Every time we strike a match we unconsciously initiate a chain reaction. The flaming head of the match raises the temperature of atoms in nearby cellulose molecules sufficiently high so that they react with atmospheric oxygen. Part of the energy released by this reaction is transferred to neighboring atoms in the match stick, and they in turn combine with other oxygen atoms. The process is repeated over and over again, and presto! the result is combustion.

On paper one can suggest many nuclear reaction cycles that ought to follow the same pattern. For example, consider the following well-known pair of reactions:



Protons released in the first reaction have sufficient energy to bring about the second reaction, and in turn regenerate fast alpha particles able to produce reaction 1 again. Why does this not happen in practice? The answer is obvious when one recalls that the nuclear physicist normally achieves but a single transmutation for every million charged projectiles he hurls at a target. The other 999,999 bullets miss the nuclear mark and dissipate their energy by producing excitation and ionization in the target material. Thus the nuclear chain reaction is effectively squelched by the law of probability.



By 1930 scientists had pretty well discounted the possibility of ever producing a nuclear chain reaction on earth. Then the discovery of the neutron served to revive their hopes for a spell. Here was a particle which, being uncharged, did not interact with the electronic structure of an atom, nor did it need the energy to surmount a coulomb barrier in order to enter a nucleus. Place a neutron in a large mass of material and it is inevitable that combination with a nucleus will be effected. Given 1000 free neutrons one can practically guarantee 1000 transmutations. Then came the discovery of  $(n, 2n)$  reactions, and the chain-reacting possibilities seemed even more promising. But investigation soon proved that these reactions were always endoergic, i.e., the two outgoing neutrons invariably had less energy than the original neutron and hence the former were unable to propagate the  $(n, 2n)$  process. Again, talk of harnessing subatomic energy died away. Then suddenly with the discovery in 1939 of (1) nuclear fission by neutrons and (2) the accompanying secondary neutrons, the idea of releasing nuclear energy on a grand scale again came to the fore. This time the dream was no will-o'-the-wisp, but the story leading to the final achievement is one of keen insight, remarkable ingenuity, infinite patience, and hard labor. In recounting this development the authors cannot hide a real feeling of admiration for that band of scientists who wrested the secret from nature, a secret so well hidden and protected by natural bulwarks as to make it appear that some superior force had in this manner guarded a most treasured possession.

Focusing our attention on uranium we recall that both prevalent isotopes  $U^{238}$  (99.3 per cent) and  $U^{235}$  (0.7 per cent) can be fissioned by neutrons. Curves *a* and *b* of Fig. 1 show qualitatively how the macroscopic fission cross section ( $N\sigma_f$ ) for each isotope varies with neutron energy. We can assume that each fission event will release, in addition to 160 Mev kinetic energy in the fission fragments and 5 Mev gamma energy, about two secondary neutrons each having an energy of roughly 2 Mev. The main problem resolves itself into making these neutrons bring about further fissions so that a continuing reaction can be established. Our problem would be extremely simple if fission were the only mechanism through which neutrons interact with uranium nuclei, but unfortunately this is not the case. The following important processes are also possible:

1. Elastic scattering.
2. Inelastic scattering of fast neutrons.
3. Radiative neutron capture by  $U^{238}$  to give  $U^{239}$ .



Owing to the heavy mass of a uranium nucleus, we can ignore the practical effect of elastic scattering. In a general way the variations in cross section with energy for the other processes are displayed by curves *c* and *d* of Fig. 1. Considering the strong  $U^{238}$  resonance capture level at  $\approx 7$  volts (there are also other less prominent resonance levels not shown), it is apparent that the only hope of achieving a chain

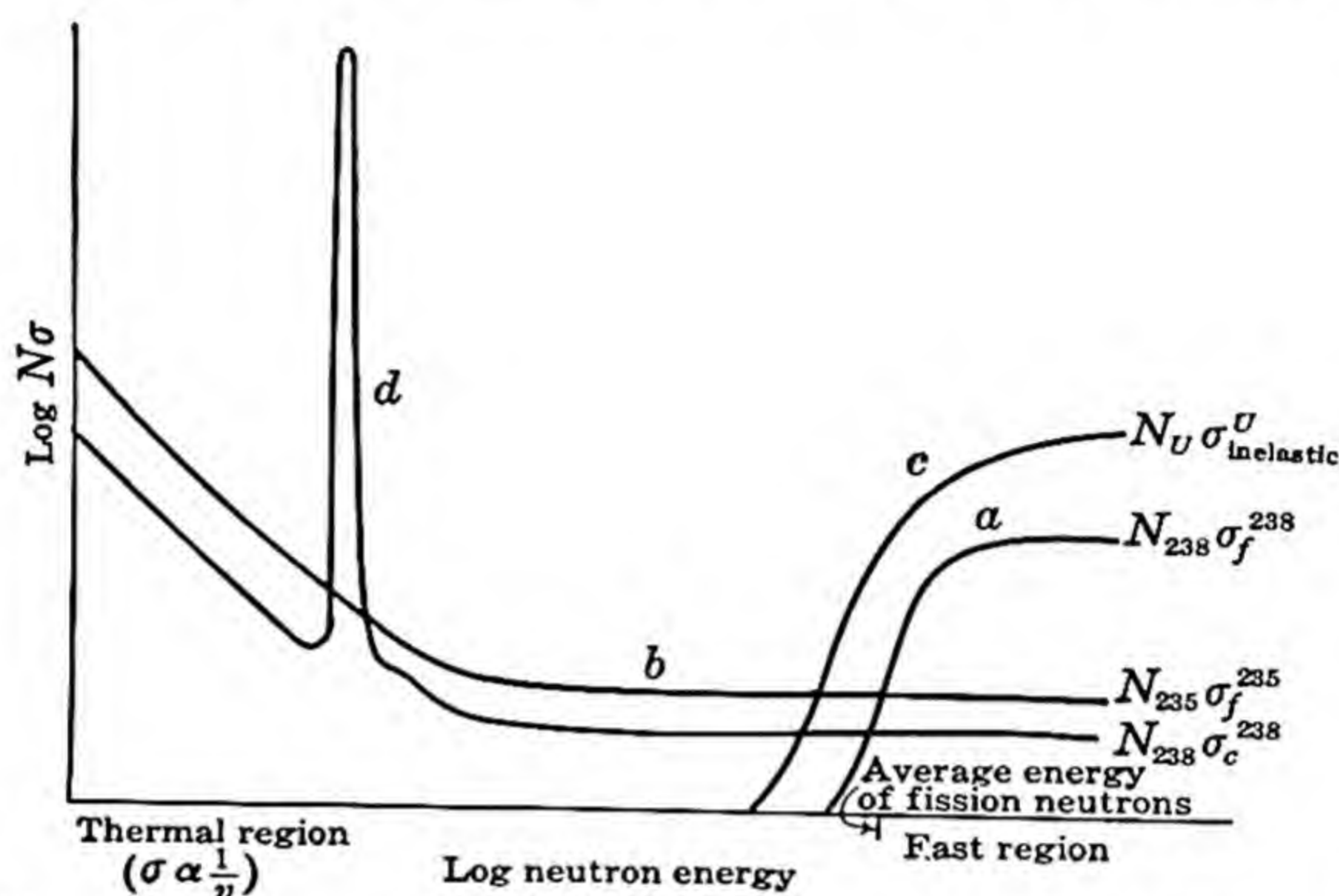


FIG. 1. These curves show in a very qualitative way how the cross sections for the interaction of neutrons with uranium isotopes vary with neutron energy. Here  $N$  refers to the number of atoms of a particular isotope per cubic centimeter.  $\sigma$  has its usual significance. Hence  $N\sigma$  expresses the probability that a neutron will produce an event of the type being considered in 1 cm of path. This product is called "macroscopic cross section." A logarithmic scale is employed so that the wide spread in cross sections and neutron energies can be presented on a single graph.

reaction with a mass of undiluted uranium metal centers around a fast neutron fission reaction on  $U^{238}$ . But this is clearly impossible inasmuch as a single inelastic scattering event always reduces the neutron energy to a point well below the  $U^{238}$  fission threshold, and  $N_U \sigma_{\text{inelastic}}^U$  is several times greater than  $N_{238} \sigma_f^{238}$ . Consequently far less than half the secondary neutrons will be able to produce fissions in  $U^{238}$ . (The same argument can be used for the case of  $Th^{232}$ .) However, there is no fission threshold for  $U^{236*}$  ( $U^{235} + \text{neutron}$ ) so this consideration does not apply to a mass of pure  $U^{235}$ ; otherwise an atomic bomb based on  $U^{235}$  would never have been possible.

It thus becomes evident that to establish a chain reaction in ordinary uranium we are forced to fall back on the  $U^{235}$  present. Since the  $U^{235}$



fission process can compete with  $U^{238}$  capture only for neutrons with energies below 7 electron volts, we are faced with the problem of transforming the fast secondary neutrons into slow neutrons without allowing them to be gobbled up by  $U^{238}$  as they pass through the 7-electron volt resonance region. The slowing-down action can be brought about by introducing large quantities of a "moderator," comprising atoms of low mass number whose function it is to soak up energy from the fission neutrons through elastic collisions, but at the same time not absorb any appreciable fraction of the neutrons present.

In the province of light nuclei only  $H^1$ ,  $H^2$ , He, Be, C, and O have the requisite low neutron capture cross sections to qualify as good moderators. Carbon in the form of graphite proved to be the most logical candidate for early "pile" (a name given to nuclear chain reactors) work.  $H^1$  and  $H^2$  have been used successfully in later piles.

A simple but clever ruse can be used for getting the neutrons safely around the 7-electron-volt resonance trap. This entails forming the uranium into small lumps and placing these on lattice points in a matrix of the moderator, instead of uniformly mixing the two components. In this way most of the slowing takes place in a region free of uranium, and only those few neutrons which happen to enter a uranium lump with 7-electron-volt energy fall victim to the strong resonance level. Another way of discerning this effect is to recognize that any 7-volt neutron which enters the U lump will be absorbed in a thin surface layer. This thin layer thus can be said to shield the remaining volume of U from resonance neutrons while allowing relatively free passage of thermals. Even with this trick our chain reaction is not guaranteed. We must recognize that not every slow neutron captured by  $U^{235}$  must of necessity produce a fission, since there may be some non-fission capture in  $U^{235}$ . Finally there is the question of neutron loss by leakage from the pile. However, leakage is a surface effect (for a sphere proportional to the square of the radius) whereas neutron production is a volume phenomenon (proportional to the cube of the radius). Hence relative leakage loss can be reduced by increasing the reacting mass. In fact for any combination of fissionable and moderating material there will be a lower limit to the size of a workable unit, owing to the leakage loss. This limit has been given the name "critical size" and is a very important characteristic of any chain-reacting system. It should be noted that the critical size can be reduced somewhat by surrounding the reactor with a so-called "reflector." The latter improves the neutron economy by scattering a portion of the leaking neutrons back into the reactor proper where



they can be used to advantage. In general the same materials which serve as moderators are used for reflectors.

Although the basic requirements for a chain reaction in uranium were fully appreciated as early as 1941, the various nuclear constants were not known with sufficient accuracy to permit a prediction as to the chance of success. Two lines of attack appeared open to those interested in prosecuting this problem:

1. A more accurate determination of the pertinent nuclear constants together with experiments designed to yield the optimum arrangement of the pile components.

2. A direct attempt to establish a chain-reacting pile, using the best data available for choice and arrangement of materials.

Although it was realized that avenue 2 promised quicker returns there was not sufficient uranium and graphite of high purity available in 1941 to permit a direct test. Consequently attention was originally concentrated on 1 until the production of pure pile materials allowed a shift in emphasis to 2 late in 1942.

Cutting through the multitude of perplexing problems associated with a nuclear chain reaction, the following question, posed by Fermi and his collaborators, was absurdly simple. "Is it possible to produce an assembly of uranium and other materials such that if the arrangement were of infinite extent the addition of, say, 100 fast neutrons to the pile would result after one neutron generation in the presence of more than 100 neutrons within the pile?" If the answer were no, then the whole idea of a chain reaction with natural uranium could be forgotten. If the answer were yes, then one could feel hopeful that the same thing might be accomplished in a system of finite size. To put things on a more formal basis the multiplication factor  $k$  was introduced into pile terminology.  $k_{\infty}$  is defined as the number of neutrons present at the end of a neutron cycle in an infinite pile divided by the number present at the start. Hence one can rephrase the above question by inquiring whether  $k_{\infty}$  can be made greater than unity.

In order to formulate an expression for  $k_{\infty}$  in terms of measurable quantities and at the same time get some notion of the physical processes involved, let us follow these 100 fast neutrons through a complete cycle. Once added to the pile these neutrons will move about rapidly, losing energy through elastic collisions with moderator atoms and inelastic collisions with uranium nuclei. However, before they fall below the  $\text{U}^{238}$  fission threshold a small percentage will cause fission and release additional fast neutrons. Thus instead of having 100 neutrons to slow down we will actually have  $100\epsilon$ , where  $\epsilon$  is a



number slightly greater than unity. For a typical U-graphite pile Fermi reports  $\epsilon = 1.03$ . We can safely ignore the possibility of straight neutron capture in this energy region by either uranium or moderator. Once the neutrons fall below the  $U^{238}$  fission threshold the slowing-down process continues uneventfully until the  $U^{238}$  resonance capture region (5 to 100 volts) is reached. Here it is inevitable that we lose some of our neutrons. Let us take account of this by assuming that a fraction  $p$  of the neutrons safely escape  $U^{238}$  resonance capture and pass on to the region of thermal energies.  $p$  is the resonance escape probability. The surviving neutrons will now diffuse through the pile, neither gaining nor losing energy, until finally they are captured, one fraction by the uranium atoms and the remainder by the moderator and various auxiliary materials which may be present. Let  $f$  represent the fraction captured by uranium.  $f$  is termed the thermal utilization factor. We now define a quantity  $\eta$  as the number of secondary neutrons (fast) produced as a result of thermal fission for each thermal neutron absorbed by the uranium. Note carefully the distinction between  $\eta$  and  $\nu$ . The latter represents the number of secondary neutrons per fission, but since not every thermal neutron absorbed by uranium causes fission  $\eta$  will always be less than  $\nu$ .

Combining the above factors, we find that our original 100 fast neutrons result in the generation of  $100\eta\epsilon pf$  new neutrons, and consequently:

$$k_{\infty} = \frac{100\eta\epsilon pf}{100} = \eta\epsilon pf$$

This equation has been titled the four-factor formula and at least in principle permits one to calculate  $k_{\infty}$  for any proposed pile. These quantities can be evaluated in a reasonably straightforward manner for a homogeneous mixture of uranium and moderator. However, the problem becomes quite difficult when the pile contains uranium in the form of lumps. Even so the theoretical group at the Metallurgical Laboratory under Wigner did some outstanding work during the war in this connection.

Fermi and his associates on the other hand utilized a direct experimental approach in their quest of  $k_{\infty}$ . The method employed made ingenious use of what is called an exponential pile.\* Essentially their procedure enabled them to infer from measurements on a relatively

\* The Germans also followed the same general approach, and in their hands these units were known as neutron-injected piles.



small pile-like structure what the multiplication factor would be for an infinitely large lattice of the same type.

The first pile of this kind consisted of a graphite cube about 8 feet on an edge and contained some 7 tons of uranium oxide in iron containers, distributed at equal intervals throughout the graphite. It was realized that this unit was too small to be chain reacting. However, a Ra-Be neutron source placed at the bottom of the pile established an equilibrium neutron density throughout the lattice structure, and this density was measured along the vertical axis of the pile. It was determined that the neutron density for such an arrangement decreased exponentially with increasing distance from the neutron source. Furthermore from such rates of decrease it was possible to calculate  $k_{\infty}$  for an infinitely large pile of the same lattice proportions. The value of  $k_{\infty}$  deduced in this fashion for the first exponential pile is not available, but a second pile constructed along the same lines gave 0.87. This was not too discouraging since both the  $U_3O_8$  and graphite used were commercial products, known to contain definite impurities. The  $U_3O_8$  was the worst offender, containing 2 to 5 per cent impurities. The supplier was able to reduce this figure to below 1 per cent, and  $k_{\infty}$  quickly rose to 0.98. By ether extraction of uranyl nitrate it was found that practically all impurities could be removed in a single step. The purified nitrate was then converted to  $UO_2$ . This oxide brought great joy to the Plutonium Project scientists because its use in an exponential pile for the first time yielded a  $k_{\infty}$  greater than 1. The actual value, obtained in July 1942, was 1.007. By this time the problem of quantity production of uranium metal had been solved. The U metal-graphite combination gave an even higher value of  $k_{\infty}$  (1.07). It is true that  $k$  for an actual pile will always be less than  $k_{\infty}$ , owing to leakage losses. However, once the pile is well above critical size and is surrounded by a reflector,  $k$  differs very little from  $k_{\infty}$ . No longer did anyone doubt the feasibility of a nuclear chain reaction. The actual operation of the first pile some months later was satisfying but not unexpected.

### First Chicago pile

Because of its historic importance a brief description of the first pile seems in order. It was assembled on the floor of a squash court under the West Stands of Stagg Field at the University of Chicago. The pile contained some 6 tons of U metal consisting of lumps spaced on a cubic lattice and imbedded in graphite. Since there was insufficient metal to accommodate every lattice point, pressed oxide lumps



were also employed. The overall structure had the shape of an oblate spheroid. Cd strips fitted into slots which ran through the pile. These strips served as control rods. In fact pushing any one of the Cd strips to the full "retard" position would keep  $k$  well below 1.  $\text{BF}_3$  neutron counters whose output registered on pen recorders were placed in and around the pile to signal the approach to the critical condition. On December 2, 1942, the pile was first put into operation by removing all but one of the Cd strips and then carefully withdrawing the last strip in small steps, always pausing between steps to observe the change in neutron level as indicated on the recorder charts. At first sight taking a pile to the critical condition might seem to be a ticklish operation. However, the following argument will prove that such is not the case if certain simple precautions are observed.

It can be shown that with a change in the effective  $k$  of a pile the thermal neutron density  $n$  varies with time  $t$  in the following manner:

$$n = n_0 e^{(\rho/l)t}$$

where  $n_0$  is the density of thermal neutrons before the change, and  $l$  represents the mean lifetime of a neutron in the pile. Now  $\rho$  (the reactivity) simply represents the net increase (if positive) or decrease (if negative) in the number of neutrons produced in the pile per neutron absorbed, i.e., the amount by which the effective  $k$  of the pile differs from 1. As long as  $\rho$  is definitely negative (control rod at "retard") the pile is subcritical and nothing exciting will happen. However, as we continue to withdraw the control rod or strip,  $\rho$  becomes less and less negative and finally a stage will be reached when  $\rho$  suddenly changes sign. With the control rod accidentally pulled out well beyond the critical point,  $\rho$  might easily assume a value as high as  $+0.005$ . Since the lifetime of a thermal-pile neutron is  $\approx 0.001$  second, this would result in an  $e^{(5)(2)} \approx 22,000$ -fold increase in the neutron density within 2 seconds. Inasmuch as  $n_0$  near the critical point is considerable, such a result would be catastrophic. Happily the above representation is not correct, for which we must thank the small number of delayed neutrons attending the fission process. Though amounting to but 0.76 per cent of the total, these neutrons are delayed on the average by about 10 seconds. This increases the true mean lifetime of a fission neutron to about 0.1 seconds. We can now tolerate a  $\rho$  of  $+0.005$ , since the neutron density after 2 seconds will be only  $e^{(0.05)(2)} \approx 1.10n_0$ . It should be emphasized that this analysis breaks down if we permit a reactivity change so great



( $\rho > +0.0076$ ) that the pile is supercritical on prompt neutrons alone.

So effective were the delayed neutrons in the first pile that with the Cd strip 1 cm out from the critical position 4 hours were required for the neutron intensity to double. Thus, in the light of experience, placing a natural uranium thermal pile in operation is indeed a prosaic chore.

The first Chicago pile was definitely of the Model T variety. It boasted no adequate shield, no means for dissipating the heat released within the uranium lumps, and only rudimentary controls. However, it did prove that a nuclear chain reaction could be achieved and provided valuable knowledge on which to base the design and operation of the Clinton pile.

### Clinton pile

This pile was built in 1943 for the explicit purpose of manufacturing a few grams of  $\text{Pu}^{239}$ , which were badly needed to test the proposed chemical separation process. The pile consists of a large graphite cube honeycombed by horizontal channels filled with uranium. The uranium is in the form of short metal cylinders contained in gas-tight casings of aluminum. These "slugs" may be slid into the channels, leaving space for cooling air to flow past. They may be pushed out the back of the pile when ready for processing and be replaced by fresh slugs. The pile is surrounded by a heavy concrete shield. Neutron intensity in the pile is measured by  $\text{BF}_3$  ionization chambers and controlled by boron steel rods that can be moved in and out of the reactor by a servomechanism. This pile was designed to operate at a power level of 1000 kilowatts\* but a better fan system and other improvements allowed the level to be raised considerably. Here it might be pointed out that theoretically a pile can operate at any power level desired. In actual operation the maximum permissible pile temperature, and thus the rate at which heat can be removed from the system, normally sets a practical upper limit. For any constant power level the effective  $k$  ( $k_{\text{eff}}$ ) is always 1. Ignoring second-order effects, a change from one power level to a higher one is accomplished as follows: First the control rod is retracted slightly. This makes  $k_{\text{eff}} > 1$  and so the neutron density, and hence the power level, increases exponentially as described previously. Once the new level is reached the

\* Each fission event releases K.E.  $\approx 200 \text{ Mev} = 3.2 \times 10^{-4} \text{ erg} = 3.2 \times 10^{-14} \text{ kilowatt-second}$ . Hence the power level of pile in kilowatts  $= (3.2 \times 10^{-14}) \times (\text{Number of fissions per second in pile})$ .



control rod is returned to its original position and the pile stabilizes at the new value,  $k_{\text{eff}}$  returning to unity

### Hanford piles

These piles, three in number, were built to produce  $\text{Pu}^{239}$ . Consequently it was necessary that they operate at the highest possible power level. This was accomplished (1) by increasing their size over that of the Clinton pile, and (2) by adopting water cooling. Water cooling is much more effective than air cooling in removing heat from the reactor, although it does raise serious piping and corrosion problems. These were satisfactorily solved, and by the summer of 1945 the Hanford piles were operating at the designed power, producing  $\text{Pu}^{239}$ , and heating the Columbia River.

As the chain reaction proceeds, the  $\text{U}^{235}$  in the pile becomes depleted. However, the concentration of  $\text{Pu}^{239}$  in the slugs increases. Because this isotope is also thermally fissionable its presence tends to counterbalance the decrease of  $\text{U}^{235}$  as far as maintaining the chain reaction is concerned. But the concentration of fission products also increases with time, and these can "poison" the pile by neutron capture. The original Smyth report reveals that in spite of a great deal of preliminary study on fission products an unforeseen poisoning effect of this kind very nearly prevented operation of the Hanford piles.

### Piles moderated with heavy water

If the laws of simple mechanics are applied to the elastic collisions of neutrons with moderator atoms, the conclusion is that the neutron loses a constant fraction of its energy per collision. This suggests the introduction of a term  $\xi$ , defined as the average loss in the logarithm of the energy in one collision. In other words

$$\xi = \overline{\ln E_0 - \ln E} = \overline{\ln \frac{E_0}{E}}$$

where  $E_0$  refers to the neutron energy before collision, and  $E$  is the energy after collision. The utility of  $\xi$  arises from the fact that it is independent of the neutron energy. Simple considerations lead to the following expression for  $\xi$ :

$$\xi = 1 + \frac{r}{1-r} \ln r$$



where  $r = \left(\frac{A-1}{A+1}\right)^2$ ,  $A$  being the mass number of the moderating material. If we know the value of  $\xi$  for a moderator we can readily calculate the average number of collisions  $\bar{N}$  a neutron must make in slowing from fission energy (2 Mev) to thermal energy (0.025 volt). This will be just the total loss in logarithm of energy divided by  $\xi$ , i.e.,

$$\bar{N} = \frac{\ln(2 \times 10^6) - \ln(0.025)}{\xi} = \frac{18}{\xi}$$

Table 1 records these quantities for several potential moderators, together with figures for the absorption cross section ( $\sigma_{\text{ath}}$ ) for thermal neutrons.

TABLE 1

MODERATOR	$A$	$\xi$	$\bar{N}$	$\sigma_{\text{ath}}$ (barns)
Hydrogen	1	1.000	18	0.30
Deuterium	2	0.725	25	0.0006
Beryllium	9	0.209	86	0.009
Carbon	12	0.158	114	0.005
Oxygen	16	0.120	150	0.0016

It is apparent from Table 1 that graphite is not the ideal moderator. Its choice for the early pile work was dictated by its availability and low neutron capture cross section. The latter consideration prevents the use of ordinary hydrogen (water) as a moderator in natural uranium piles. Heavy water ( $\text{D}_2\text{O}$ ) on the other hand seems a logical candidate on both counts and to date two piles have been built employing this moderator.\*

The first was constructed at the Argonne Laboratory (Chicago) in 1944 and, owing to the efficient moderator, is of very small size. Its rated power level is 300 kilowatts, and the thermal neutron flux in the center is  $10^{12}$  neutrons/cm<sup>2</sup> second. The general characteristics of this pile differ somewhat from those of comparable graphite piles. It shows

\* On the basis of some incomplete experiments the Germans decided in 1941 that a chain reaction could not be established in natural uranium with graphite as moderator. Therefore they turned their attention to large-scale production of  $\text{D}_2\text{O}$  as the only feasible means of obtaining a moderator for a pile. This proved to be a herculean task and delayed the German atomic energy project by at least three years. Except for this, German scientists would almost certainly have achieved a successful nuclear chain reaction.



sudden fluctuations ( $< 1$  per cent) in power level due to the formation of bubbles in the water. This serves to point up a factor which must always be given due consideration in the choice of pile materials; namely, the possible damaging effect on such by the intense radiations present. This pile cannot be shut down so completely nor so rapidly as a graphite pile because of photoneutrons produced in the heavy water by delayed gamma rays.

The other heavy water pile was built by the Canadians at Chalk River, Ontario, and is considerably larger than the Argonne unit.

### Clinton power pile

There has been much speculation regarding the use of chain-reacting piles for commercial power production. Although existing piles produce vast quantities of heat, this energy is liberated at such a low temperature ( $\leq 150^\circ \text{C}$ ) that it cannot be used efficiently in heat engines. Since the thermodynamic efficiency of a heat engine is  $(T_h - T_c)/T_h$  ( $T_h$  being the absolute temperature of the working fluid as it enters the system, and  $T_c$  the exhaust temperature) it is imperative that the pile release its energy at the highest feasible temperature level. A pile is virtually unlimited in the temperature it can produce, as witness the stellar temperatures developed in the atomic bomb. However, the melting points of structural materials set a usable upper limit of a few thousand degrees for  $T_h$ .

At the present time it is not easy to predict just when the first pile for the production of useful power will go into action. It is not far from the truth to say that the jump to a high-temperature power pile today represents as great a step into the unknown as did the construction of the first Chicago pile in 1942. However, in several nations the problem is being prosecuted with vigor, and it seems safe to say that nuclear power will become a reality within the next decade. We present in Fig. 2 the sketch of a possible power pile designed to operate at temperatures up to  $1000^\circ \text{F}$ . At one time this pile was slated for construction at Oak Ridge, but this program has been canceled.

When it is finally built, the first power pile may show little resemblance to this preliminary design. Nevertheless it is of interest to consider the basic features and general mode of operation of this pile as representing the latest information to be made public on the subject. On the basis of electric-power generation practice there is nothing revolutionary in this proposal. Every element present in a conventional power station is found here. The nuclear reactor simply sup-



plies the necessary energy in the form of heat which is converted to mechanical and electrical energy in the usual way. It is obvious that the high temperature of operation imposes stringent structural requirements on the choice of materials and will also modify the nuclear behavior of the chain reaction to an appreciable extent. Once the unit has been used, direct access to the reactor for purposes of maintenance

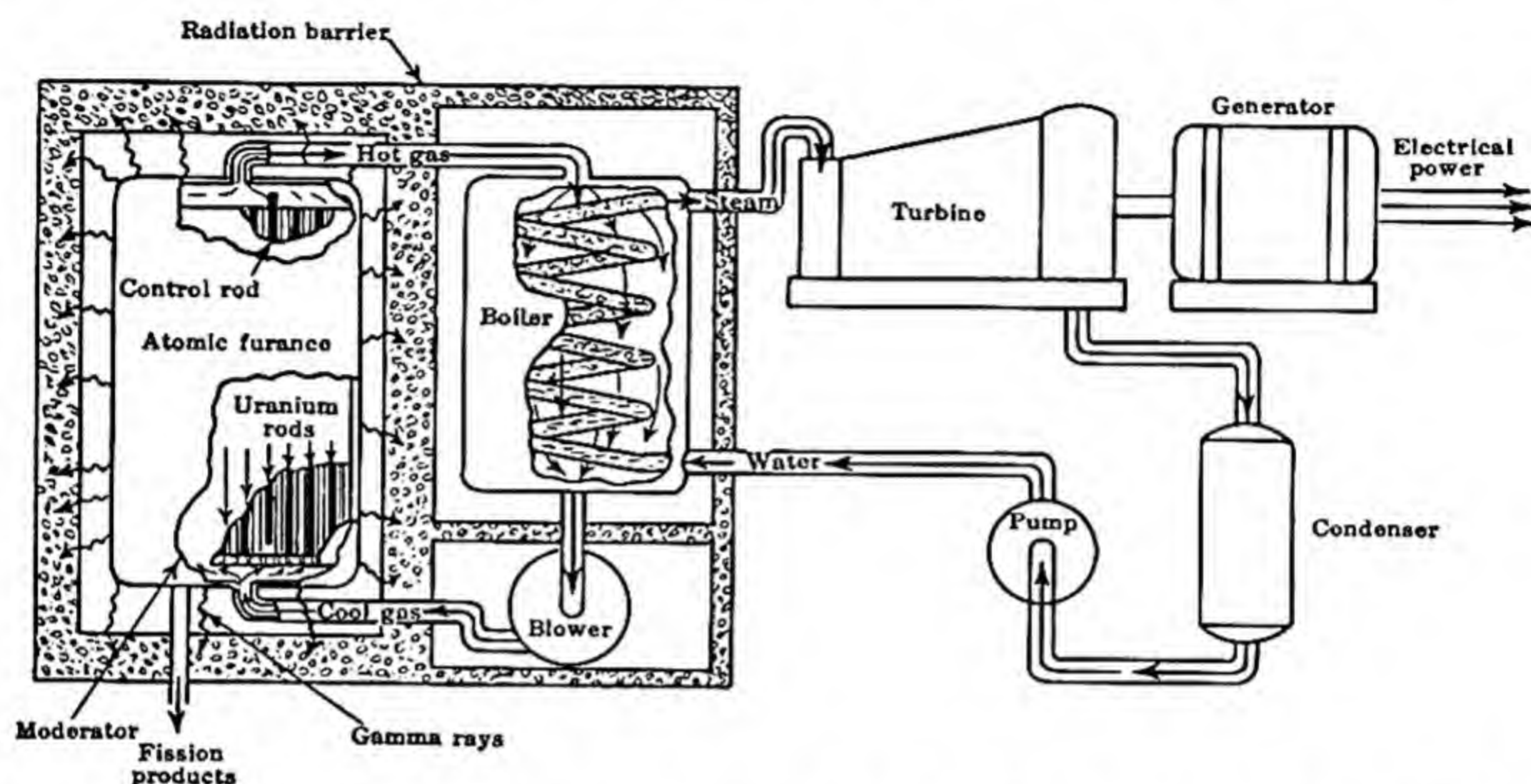


FIG. 2. This sketch, released jointly by the Manhattan Engineer District and The Monsanto Chemical Company, shows the general features of the power pile once planned for construction at Oak Ridge. The heat released in the uranium rods by the fission chain reaction is conveyed to the boiler by a circulating gas kept under high pressure. Except for the novel energy source conventional power station practice is followed. Note the heavy radiation shield which surrounds the pile and the lighter shield around the boiler. The latter shield gives protection against any activity which may be present in the cooling gas. Radioactivity in the steam will be negligible. Despite the simple drainpipe pictured, removal of the fission products will entail reprocessing the uranium rods outside the pile.

and repair will be well-nigh impossible owing to the high radiation level present even after the reactor is shut down. Consequently durability of the component parts must be given special consideration. For all these reasons much preliminary work is indicated before a satisfactory design can be evolved.

With the exception of the Clinton power pile, all previously discussed chain reactors utilize natural uranium as the nuclear fuel. This means they rely primarily on fission of  $U^{235}$  by slow neutrons, and for that reason they are termed thermal piles. Each is of the lattice type since no homogeneous mixture of natural uranium and a moderator can



sustain a chain reaction (with the single exception of U plus  $D_2O$ ). However, once we admit the possibility of obtaining U enriched in isotope 235, or pile-produced  $U^{233}$  or  $Pu^{239}$ , the above limitations may be cast to the winds. We are no longer restricted to lattice arrangements, and we may consider the utilization of intermediate or even fast neutrons to propagate the fission chain. Already a small homogeneous reactor known as the "water boiler," fueled by a water solution of uranium salt enriched in  $U^{235}$ , has been constructed at Los Alamos. In addition, a controllable Pu pile functioning on fast neutrons has also been operated at the same location.

Thus it becomes apparent that future piles will consist of several different types. One classification scheme identifies the pile according to the energy of the neutrons which cause the majority of fissions. On this basis we can imagine piles ranging all the way from thermal to fast. The category to which any pile belongs is determined by the amount of moderator present, i.e., a thermal pile contains large amounts of moderator whereas a fast pile contains no intentional moderating material, although inelastic collisions with the heavy atoms present reduce the neutron energy somewhat. The very fact that various classifications find ardent supporters among pile scientists leads us to suspect that no one type can claim superiority over the others on all counts. Just as the pony, the work horse, and the thoroughbred can each justify his existence as a member of the equine family, so can each pile boast certain advantages that merit serious consideration. To gain an understanding of the situation let us compare briefly thermal and fast piles, using as criteria the following important considerations: (1) critical size and specific power output; (2) neutron economy; (3) reprocessing of fuel.

1. *Critical Size and Specific Power Output.* From Fig. 1 it is evident that the fission cross section for  $U^{235}$  is much greater for thermal than for fast neutrons. This implies that the amount of fissionable material to build a critical thermal unit will be considerably less than is required for a fast pile, which could mean that enough fissionable material to construct five fast piles might be adequate for ten of the thermal variety. Though more fissionable material is required, the absence of a moderator results in the fast reactor having a much smaller volume than its thermal counterpart. As it turns out this small size is the major disadvantage of a fast reactor, for it seriously limits the rate at which heat can be withdrawn. If one tries to increase the reactor surface through voids or dilution with heavy atoms,



the required amount of fissionable material soon becomes prohibitive. Consequently the specific power level, i.e., the permissible heat output in kilowatts per kilogram of fissionable material is relatively low for a fast pile. On this basis a 100,000-kilowatt pile of this type would necessitate a much greater initial expenditure for nuclear fuel than would a thermal unit with the same power rating.

2. *Neutron Economy.* Since the ratio of (moderator plus coolant plus structural atoms) to (fissionable atoms present) decreases greatly as we proceed from a thermal to a fast pile, we naturally would expect the relative neutron losses to be least in the fast pile. In addition, no element shows strong capture resonances for fast neutrons. Therefore small amounts of impurities such as boron and even fission products will not drastically impair the fast pile neutron economy as they do for thermal piles. This means that fast piles should be more efficient than the other types in providing excess neutrons for other uses. This brings us to the subject of "breeding." Whenever a fissionable atom in a pile captures a neutron and divides, it is necessary that we expend one of the secondary neutrons to bring about another fission if the chain reaction is to be maintained. Theoretically all the remaining secondary neutrons could be channeled into "fertile"  $U^{238}$  or  $Th^{232}$  nuclei to create new fissionable atoms. It is clear that if  $\nu$  were greater than 2 we might conceivably produce more  $Pu^{239}$  or  $U^{233}$  atoms than the  $U^{235}$  consumed. This would amount to true breeding and would provide a means of increasing our stockpile of fissionable material and enable us to utilize all the U and Th on earth to release nuclear energy rather than being limited to  $U^{235}$  alone. The Hanford piles represent a step in this direction, but the  $Pu^{239}$  produced there does not compensate for the  $U^{235}$  burned; so this is breeding in a rather limited sense.

3. *Frequency of Reprocessing.* It would be nice if we could simply introduce our fuel rods into a pile and then forget them until all their fissionable atoms had been burned. Usually this will not be possible because of the build-up of parasitic fission products. As a consequence it is necessary to remove the fuel units occasionally for reprocessing by chemical means. Each trip through the processing plant is certain to result in some loss of precious material, and the frequency of reprocessing must be taken into account in selecting a pile type. Some proposals have been made relating to liquid piles. Here the fission products might be removed more or less continuously, thus simplifying the problem of reprocessing.



### Future trends in pile design

Having surveyed in a qualitative manner the two main pile types, let us conclude this chapter by speculating briefly on possible future trends in pile design.

We have seen that the earliest piles were necessarily heterogeneous in nature. However, it was also noted that the availability of enriched fuel made it possible to postulate homogeneous arrangements, and we saw that such reactors, especially those of a fluid nature, offered certain definite advantages. In addition to the obvious reasons for preferring a fluid reactor (ease of fission product removal, etc.) the novel suggestion has been made that the fluid fuel be circulated through a heat exchanger to eliminate the need for a separate coolant. At first this seems an attractive idea, but on closer inspection a number of problems arise. A greater quantity of fissionable material is then needed since that in the circulating system is of no help in maintaining the chain reaction. Some of the delayed neutrons would be released outside the reactor and thus control would be more hazardous. Finally both reactor and heat exchanger would have to be heavily shielded for radiation protection whereas normally the latter requires only a light shield. These factors seem to place the circulating pile in a rather unfavored position.

The heterogeneous pile still has its supporters, mainly by reason of its engineering advantages. Until adequate supplies of concentrated fuel are available we are sure to see reactors of this nature being built. At the moment it is impossible to say which arrangement will be more popular a decade hence.

Except for a few research piles it is a foregone conclusion that future reactors will produce large quantities of heat at a high temperature level. Thus the problem of heat removal is a crucial one, and the manner of cooling the pile gives a further means for grouping piles into several categories. In addition to the scheme for circulating the fuel there are three proposals for cooling a pile. They include:

1. Liquid metal coolant.
2. High-pressure gas coolant.
3. High-pressure water coolant.

The first method has much to recommend it and is the only feasible plan for extracting heat from a fast pile. In any event the fuel would have to be jacketed since U and Pu react with liquid metals at high temperatures. For thermal piles Bi, Na, or a Na-K alloy appear to be promising coolants. Hg or Bi would be satisfactory for a fast pile.



With reference to the second method, gases are rather inefficient as heat transfer media, even at high pressures. However, this deficiency may be largely offset by permitting the use of simplified fuel rods. In fact this was the cooling method planned for the first power pile (See Fig. 2).

It is clear that at the proposed operating temperatures of power piles a cooling system which employed water would have to be maintained at very high pressures.

Figure 3 presents a diagram which attempts to summarize at a

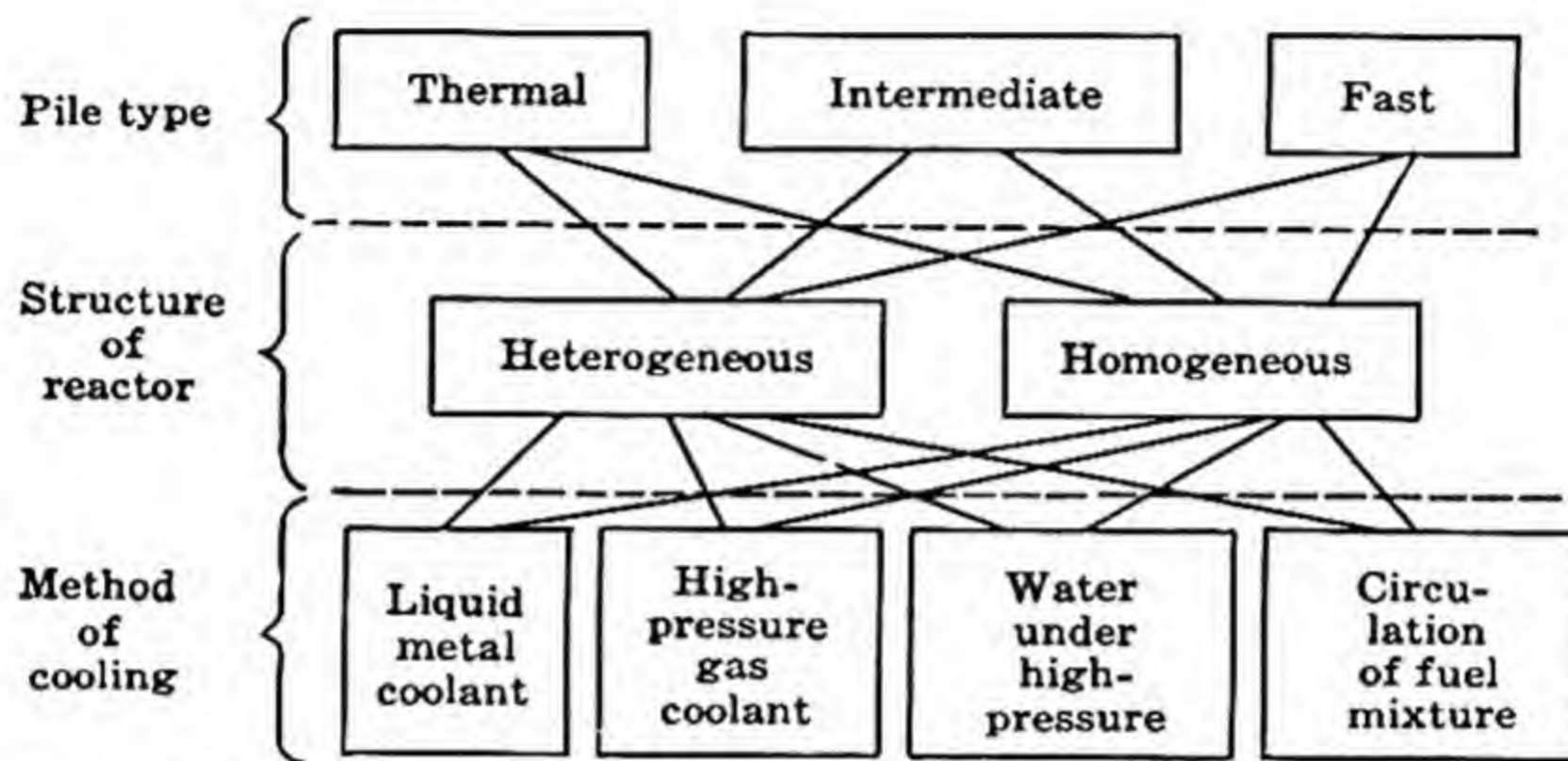


FIG. 3. The interrelationship between various types of chain reacting pile.

glance the general lines along which present pile design is going forward. When one appreciates the many engineering materials available for each component part and the various ways of arranging these, it becomes evident that the number of different piles that can be visualized is practically infinite. Consequently it would appear futile to discuss any one specific design as representative of future power piles. Both nuclear and conventional physical and chemical properties must enter into the choice of pile materials. Future technological advances will certainly expand the list of possible structural materials.

### Reactor program of the Atomic Energy Commission

In May 1949 the United States Atomic Energy Commission announced plans for the construction of four new types of reactors. (The term *reactor* is now generally preferred to *pile* for describing a nuclear chain-reacting unit.) They are:

1. A materials testing reactor which, as its name indicates, will be used in the study of materials to be employed in building reactors. Fuel elements will be U enriched in  $U^{235}$ . (Construction plans being



prepared jointly by Argonne National Laboratory and Oak Ridge National Laboratory.)

2. A Navy reactor designed as a land-based prototype of a reactor for use in propelling naval vessels of appropriate types. Fuel elements will be U enriched in  $U^{235}$ . (Design by Argonne Laboratory. Construction by Westinghouse Electric Corporation.)

3. An experimental "breeder" reactor designed to operate with high energy neutrons. This will be used to explore further the possibilities of breeding, i.e., producing more fissionable material than is consumed in the operation of the reactor and also of power production. Fuel elements will be  $U^{235}$ . A liquid metal coolant will be used. (Design by Argonne Laboratory.)

4. An experimental "breeder" reactor designed to operate with neutrons of intermediate energy and to explore their possibilities for breeding, as well as to produce usable power. A liquid metal coolant will be used. (Construction by General Electric Company.)

The AEC has announced more recently that project 4 has been postponed for the present, owing to the pressure of more important work.

In order to facilitate the reactor program a field station has been established at Arco, Idaho. The materials testing reactor and the Navy reactor will be built at this site. Facilities will also be set up at Arco for the chemical processing of reactor fuel elements and the recovery of useful material as well as for the concentration and handling of fission products from the reactors.

A long-range project to study the possibility of nuclear energy for the propulsion of aircraft (NEPA) was established at Oak Ridge in 1946. It is being managed by the Fairchild Engine and Airplane Corporation under the supervision of the Air Forces and in cooperation with the Atomic Energy Commission.

Finally it should be mentioned that studies are also being carried out on the design of a simplified research reactor. Such a unit designed primarily for research purposes and the training of new technical people in this field should cost much less than the aforementioned reactors.

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## 12. Nuclear Theory and Cosmic Rays

*What we know here is very little; but what we are ignorant of is immense*—A quotation from Laplace seen on the bulletin board of a famous radiation laboratory.

It is partially true that we have selected the title to this book with malice aforethought, because we would not have to expound on the theory of the nucleus. In spite of this we cannot bring ourselves to leave the reader without some idea of how the nature of the nucleus itself appears to the theoretical physicist. The reader may note there a little canny wording, for we have not made any claim about a description of the nucleus itself—only how it appears at present to the theorist. It is so likely that all the discoveries necessary to the founding of an adequate nuclear theory have not yet been made that any theory must be accepted as provisional and left at that.

Before we consider the nucleus, a glance at the atom as a whole is most instructive. It is generally described more or less as follows: electrons are moving according to the rules of quantum mechanics in the field of one another and of the charged nucleus, which nucleus is characteristic of the element. If the motion of the electrons is correctly calculated according to the required rules, supposing that the only type of force operative is the coulomb force between charges, then a very satisfactory theory of the atom results. The excellence of the agreement between prediction and findings based on this approach gives us great confidence in the validity of quantum mechanics and leads us to hope that it may be valid also in a region of nature which is several orders of magnitude smaller than the whole atom. This, however, is no more than a hope.

Now the nucleus itself offers a much harder problem in calculation for two reasons. The first, and more obvious, is that we cannot make the assumption that the only force operative is the coulomb force, because, if it were, the protons would blow themselves apart immediately. The second is that there is no such simplification as having the



great majority of the force field due to a nucleus at the center of the atom with light electrons rotating around. Instead of this simplification we have the necessity of considering each particle in the nucleus to be as good as another, and we are therefore presented with a complicated "many-body" calculation for any but the simplest nuclei.

### Nuclear forces

The first of the difficulties above is intriguing as it sets us the task of determining the characteristics of some new forces of nature. Accordingly we can consider in the first place what evidence we have about the forces which operate in the nucleus, namely, those between neutrons and protons, neutrons and neutrons, and protons and protons. The neutron-proton "interaction" can be studied by considering the nature of the deuteron. The deuteron is a combination between a neutron and a proton. If the force between a neutron and a proton is very large the combination will be extremely stable, which means that a considerable amount of energy will be expended in separating the two. If the force is feeble, little energy is needed. Thus, by measuring the *binding energy*, some information regarding the neutron-proton force can be found. In nuclei this binding energy is very large, amounting to several Mev, and as the theory of relativity requires an equivalence between mass and energy the binding energy of a deuteron can be found by measuring the mass of the deuteron and finding the difference between the separate masses of the two components of the deuteron and the mass of the combination. Thus the relative mass of the deuteron is 2.0147, while the sum of 1.0081 and 1.0089, the separate masses of the proton and neutron, respectively, is 2.0170. The difference is 0.0023. In actual grams for a single deuteron this is  $0.0028 \times (1.6 \times 10^{-24})$ , and if we multiply by the square of the velocity of light to get the equivalent in ergs, according to the theory of relativity, we obtain  $4.0 \times 10^{-6}$  erg. In Mev this is 2.1.

In this way we can obtain an experimental value for the binding energy between the stable combination of a neutron and a proton. Now we are presented with the problem of using this to give a definite meaning to the interaction between the two. Remember that we are only at the very beginning of our work and that we expect no more than a part of the story from this one line of evidence. With this in mind it is not too difficult to see that the theorist represents the force between the neutron and the proton by a *potential field* as in Fig. 1, either A or B. The meaning of this form of representation is that we consider the motion of one particle in the field of the other and suppose



that the potential energy due to this field can be represented by either a well or an indentation as shown in the two parts of the figure. Now the particle we consider will have a certain amount of kinetic energy as well as potential energy and will move so that its total energy is a constant. This total energy may be represented by a line as at  $G$ . The binding energy, the energy needed to separate the two particles so that they do not influence one another at all, is not the energy of the depth of the whole potential well,  $OD$  in the figure, because the

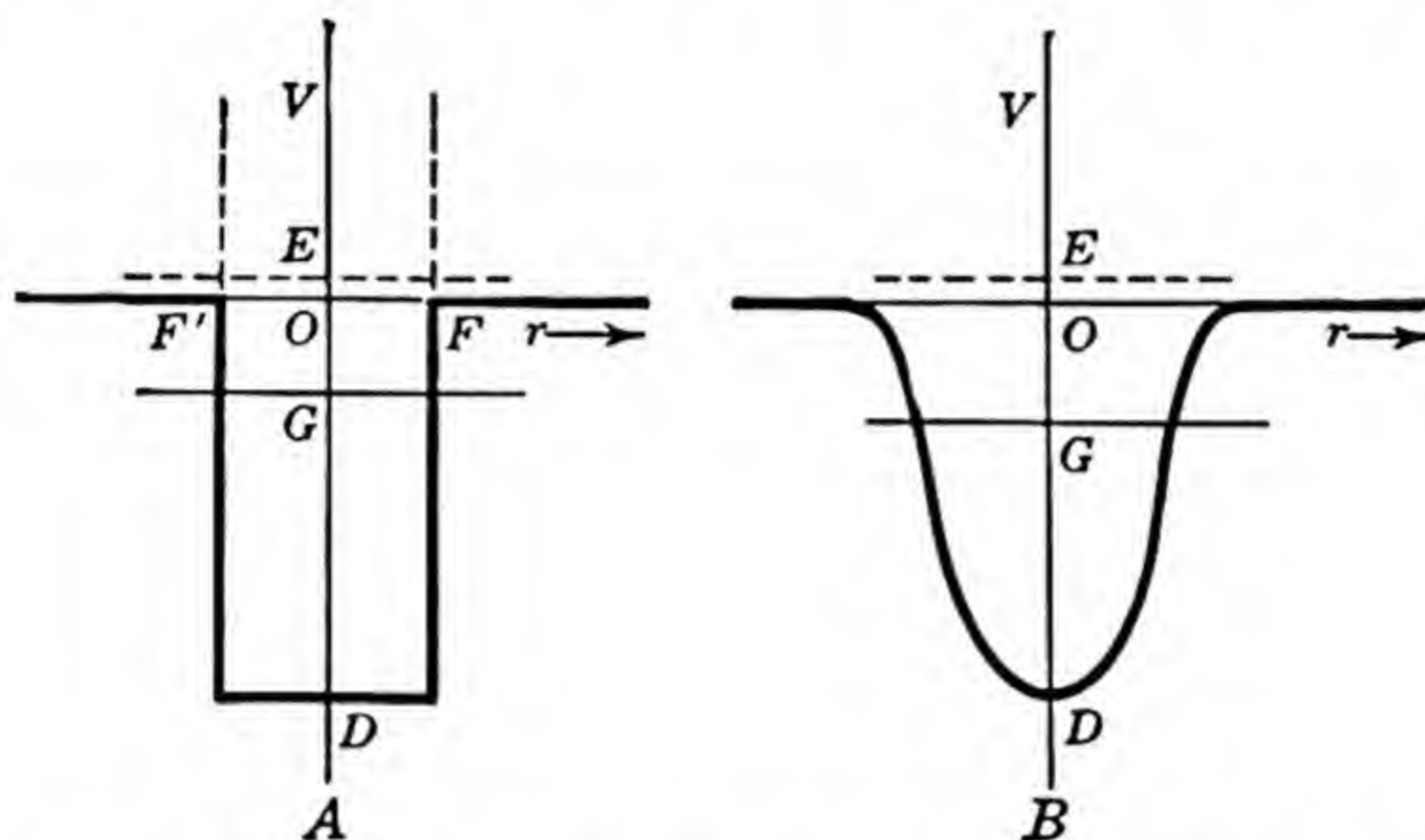


FIG. 1. Two ways of representing the potential field due to a force between a neutron and a proton, the force being that which makes possible the existence of a deuteron. Since the proton and neutron are in motion they possess kinetic energy, and the binding energy of the deuteron is determined by the position of the energy level,  $G$ , in the potential well.  $GD$  represents the total energy, kinetic and potential, inside the well, and  $OG$  the binding energy.

kinetic energy of the particle helps this separation, but the amount of energy represented by the potential  $OG$  in the figure. We then have the experimental fact that  $OG$  is 2.1 Mev for the deuteron.

To use this information that the binding energy is 2.1 Mev we have to devise potential fields which will give the right value. If we choose the well type of field we can describe the well by two magnitudes, the radius  $OF$  and the depth  $OD$ . Using this description we proceed as follows: We know from quantum mechanics that the motion of the neutron or proton is governed by a wave equation and that this has certain solutions which have physical meaning. These solutions can be obtained only for certain values of the energy difference,  $GD$ , and, if we can find a value of the two magnitudes  $OF$  and  $OD$  which make the first of these definite energy values (eigenvalues) such that  $OG$  is 2.1 Mev, we have obtained as much information as possible from the experimental value of the binding energy. It is not surprising that, with-



in a range of values, we can choose several sets of radii and depths; we could hardly hope that one fact about one nucleus would give the complete story about this new type of force. We do, however, learn that the range of values requires the radius of the well to be about  $3 \times 10^{-13}$  cm and the depth about 20 Mev. The force is thus confined to an extremely small radius and is then of enormous size.

Before we continue to see what further we can learn about the nature of nuclear forces let us consider what types of forces we already understand and what factors affect these forces. The first type is the simple force, such as a coulomb force, or gravitation, which is simply a definite function of the distance between two particles. Adding more charge (i.e., more electrons or protons) simply increases the force proportionally. A second type is commonly found between atoms and has no counterpart in so-called ordinary forces. This force can be explained only by quantum mechanics and arises in a manner related to the peculiar nature of the waves associated with matter. In bare essentials quantum mechanics requires that the description of the motion of a particle be in terms of a wave equation in which the wavelength of the waves *depends on the energy* they have associated with them.

To be more precise, if a particle moves with a total energy  $E$ , in a potential field represented by  $V$ , the wavelength of the waves describing the motion is  $h/[2m(E - V)]^{1/2}$ . This means that, if a particle has operating on it some influence which causes the wavelength of these waves to change, it must suffer some change of  $(E - V)$ , and this cannot be done without the existence of a force. If, then, we find that the predictions of wave mechanics lead us to assert that there is a change in the wavelength of the waves describing the motion of a particle we are compelled to postulate the existence of a force.

Now consider two identical atoms each containing electrons, or, to simplify it more, consider two hydrogen nuclei, reasonably close to each other, with only one electron between them. We may represent each nucleus as a field of attraction, and for ease of drawing we may represent this field as a well. Then, as in Fig. 2, we have these two wells close together; and like the deuteron the electron has a definite

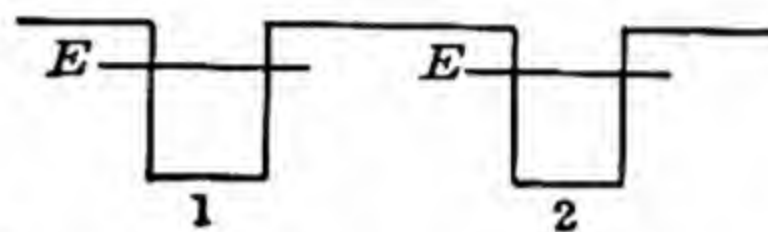


FIG. 2. Representation of the fields of two hydrogen nuclei with one of the energy levels of an electron indicated as at  $E$ . Since the wall between the two atoms is not infinitely thick there exists a chance that an electron in atom 1 find itself in atom 2. This fact requires a change in the wavelength of the waves describing the motion of the electron and gives rise to an "exchange" force.



energy  $E$  in each atom. The reader will recollect that there is always a finite probability that a particle can "leak" through any but an infinitely thick wall, and noticing that the wall between atom 1 and atom 2 is not infinitely thick he will see that if at any time the electron is in atom 1 it has a finite chance of being in atom 2. This conclusion is, on the surface, harmless enough—there is no reason why an electron should stay near one hydrogen nucleus when there is an alternative nucleus near by; but when it is followed up correctly it is found that the possibility of this "exchange" requires that the original wavelength of the waves describing the motion of the electron is altered. The amount of alteration depends, naturally enough, on the thickness of the wall separating the two atoms, which depends on their separation, and the alteration requires that a *force be exerted by one atom on the other*. This force is strong enough to make the hydrogen molecule ion reasonably stable in spite of the repulsion between the two protons, which is not balanced by any coulomb force of attraction. Such forces are called *exchange forces*. This explanation may or may not appeal to the reader, but it is designed to bring out one special feature of such a force, namely, that it is confined to a *pair* of hydrogen atoms. The proximity of a third hydrogen nucleus would undoubtedly affect the nature of the force, but not considerably, since it is only the fact of exchange between two atoms that causes the force. This type of force differs considerably from coulomb forces in that it has the feature of *saturation*.

With this preliminary discussion we can see where we may next look for information about nuclear forces. We may study the binding energy of more complicated nuclei, containing many particles, and observe whether the binding energy increases proportionally to the square of the number of particles as it would if forces acting were of the unsaturated type, or to the number itself if of the saturated type.\* We quote the binding energies (difference between the masses of the neutrons and protons and that of the atom itself) for the first twelve stable nuclei. Figures are expressed in Mev.

H <sup>2</sup>	2.1	He <sup>3</sup>	7.6	He <sup>4</sup>	27.6	He <sup>5</sup>	unstable	Li <sup>6</sup>	22.2
Li <sup>7</sup>	29.1	Be <sup>8</sup>	unstable	Be <sup>9</sup>	48.3	B <sup>10</sup>	54.5	B <sup>11</sup>	66.3
C <sup>12</sup>	82.3								

\* If the attraction depends simply on the number of constituents, the energy will depend on the number of pairs we can form. This is  $N(N - 1)$ , which is roughly  $N^2$ . If there is saturation a pair can be taken only once, and so the number of pairs effective is  $N/2$ , which is proportional to  $N$ .



A continuation of these figures shows that the increase in binding energy is reasonably linear, so that the type of force we expect in the nucleus is the type which can be saturated and which would be of the exchange type. Now if we look more closely at the figures above we see that as far as the first three nuclei are concerned we are not so justified in dismissing the ordinary type of force. Thus the number of pairs in  $H^2$  is one, and the binding energy is 2.1. The number of pairs in  $He^3$  is three, and the binding energy is remarkably close to three times 2.1. In  $He^4$  the number of pairs is twelve, and the binding energy, though not exactly twelve times 2.1, is still not far from it. It is after  $He^4$  that the saturation feature begins to show up. *There is thus something inherent in the forces which requires that the saturation occur after four particles have been placed together.*

Before we continue this discussion of nuclear forces it is as well to look at all the known properties of elementary particles, particularly the neutron and proton. The obvious properties of mass and charge we can pass over; the property of *spin* requires consideration. To the chemist this property of spin is commonplace. He will have been waiting for it to be mentioned since he knows its importance in determining the nature of the elements. To those who are not so familiar with the idea we can say that in order to describe the behavior of an electron in an atom we are compelled to use four quantum numbers, three of which are related to the motion of the electron in the field of the atom and the fourth, the spin, is related fundamentally to the electron itself. It is a quantity which determines structure. This does not mean that there is any real meaning to be attached to the word "spin," in the sense of rotation; it means rather that the spin of a particle determines, among other things, in what condition we are likely to find it. The spin of both neutron and proton is  $\frac{1}{2}$ , which means that each contributes an angular momentum  $\frac{1}{2}(h/2\pi)$  in some vectorial addition of angular momenta. The fact that this spin has this value means that both these elementary particles obey the Fermi statistics, requiring that only one particle have a particular complete set of four quantum numbers.

The application of this knowledge about spins at once sheds light on the saturation process and on the forces themselves. The fact that saturation occurs when there are four particles in the nucleus correlates perfectly with the fact that two neutrons can occupy a given quantum state, one with the spin one way and the other with opposite spin, and also two protons in the same way, making a total of four particles. The next particle, whether it be neutron or proton, is com-



pelled by the operation of the Fermi statistics to go into a different level of energy, *a fact which prevents exchange forces from operating at all*, and the result is that no stable nucleus of five particles exists. If two particles are added there is some binding and it is possible to have stability. We therefore learn that forces operate only between particles in the same quantum state. We also learn rather more. In the first place these forces must not depend very greatly on the direction of the spins of the two particles relative to one another, for if we supposed that forces occurred only for parallel spins there would be no tendency for  $\text{He}^3$  to have so great a binding energy since whatever is added to a deuteron must, by the operation of the Fermi statistics, have an opposite spin to the two previous particles which have like spins by our force requirement. In the second place we see that the forces between protons and protons, neutrons and neutrons, and protons and neutrons are much alike or the binding energy of  $\text{He}^3$  would not be three times that of the deuteron and the binding energy of  $\text{He}^4$  would not be twelve times.

This reasoning may seem so complicated to the reader that we may as well pause to summarize a little. We have found that new forces are necessary in the nucleus. These forces operate over an exceedingly small range of distance; they are of the "exchange type" in that they show saturation; they operate only between particles in the same quantum state; and they are not greatly affected by the direction of the spin of one particle relative to one another.

With this much information we can begin to look around for other lines of progress. We look to that favorite method of study of the nucleus, the scattering of particles by nuclei. Better yet, we propose to study the scattering of elementary particles by one another, and this can be done in two of the three possible cases. We can study the scattering of neutrons by protons and of protons by protons, but the third we cannot yet do. The scattering of neutrons by neutrons requires that we build a container for neutrons, and that has not yet been done. By far the best information exists for proton-proton scattering, and we can therefore treat it first. The first evidence for the action of nuclear forces in the scattering of protons by hydrogen was obtained by White, using a cloud chamber to observe forked tracks due to collisions between protons and hydrogen gas in the chamber. He was able to analyze his data to show that the manner of scattering required a force at close quarters between the two protons concerned in the collision. The number of tracks he observed, however, though sufficient to show the reality of the effect of proton-proton forces, was not



enough for a real idea of their nature to be obtained. The later work of Tuve, Hafstad, and Heydenburg, in a series of studies which is perhaps the most satisfying in the whole subject of nuclear physics, shows that as the energy of the protons from an electrostatic generator is varied the manner of scattering changes from that to be expected according to a coulomb force to one which corresponds to a sharply varying nuclear-type force. Their experiments, together with later work by Herb, Kerst, Parkinson, and Plain, are very thorough and permit an accurate determination of both the breadth and depth of the potential well due to proton-proton forces. When this is done it is found that although the force between these two like particles is not so great as the force between neutron and proton it is not very much less. This agrees with the conclusion already mentioned in the last paragraph but one.

Although the experiments so far described show definitely the existence of a force between two protons and give some idea of its magnitude, they do not say anything about the cause of the force. The search for this cause is now perhaps the major driving aim of nuclear physics. One approach is to increase the energy of the protons which are scattered and see whether there is, in addition to scattering explained by a single well, some dependence on angle or energy which cannot be so explained. This added feature is then something which can be tested against various suggestions for the cause of the force in the first place. So far experiments accurate enough to permit a real test of hypotheses have not been carried out, although the energy of the scattered protons has been considerably raised. No fully completed data have yet come in from protons accelerated by the large cyclotrons which have gone into action. The results of this work will have the most profound effect on nuclear theory.

The scattering of neutrons by protons is equally important. It is harder to carry out because collimated beams of neutrons of high energy are hard to obtain. The best approximation to the required directional beam has been obtained in the large Berkeley cyclotron, where 200-Mev deuterons produce neutrons in the target which are approximately 100 Mev in energy and confined to a small solid angle in the direction of the original deuterons. These neutrons have been used to study the angular distribution of recoil protons from hydrogen, and it has been definitely shown that the exchange process does indeed take place. This is seen because too many protons are observed at low scattering angles, unless it is supposed that some of the protons were originally neutrons which have changed identity and become



protons. The results do not, however, require that the entire interaction be of the exchange type, and more work is clearly necessary to show what part exchange has in nuclear forces.

### Spin and magnetic moment

One question which immediately arises is whether the forces between elementary particles depend only on the positions of the particles or whether orientation also is a factor. Each elementary particle possesses a spin, and it is therefore readily possible that the force between any pair depends on the relative direction of their spins. That this is so for neutrons and protons has been shown in several ways. It was first suspected to be so when it was found that the absorption cross section for slow neutrons in hydrogen did not agree with the scattering cross section. This was explained by Breit and Wigner as being due to the existence of a nearly stable form of the deuteron in which the spins of the two elementary particles are opposite, whereas in the normal form they are in the same direction. The energy difference due to this is 2.1 Mev out of perhaps a 20-Mev total. It was further proved by Brickwedde, Dunning, Hoge, and Manley, who showed that scattering of slow neutrons by molecular hydrogen was markedly different if the hydrogen was in the *ortho* form, where the protons forming the hydrogen nuclei have spins in the same direction, from the *para* form, where the spins are opposite. Improved measurements by Alvarez and Pitzer and by DeWire, Sutton, and a group at Los Alamos have shown this effect more clearly and give a very high ratio for the two cross sections. It seems definite that the range of the force between two particles of opposite spin is less than that between particles with spins in similar directions.

An even greater complication has been introduced by the recently developed precision measurements of magnetic moments. The process of abrupt transition used by the atom makes possible a technique of observing any quantity which causes that transition, provided the transition itself can be detected. Rabi first exploited this idea by a method which has permitted the ultimate in precision to be applied to the measurement of nuclear magnetic moments. If a nucleus is subjected to an oscillating magnetic field and if it possesses a magnetic moment, this field will tend to reorient the nucleus. In Rabi's experiment the nucleus was first held in orientation by sending it through a fixed magnetic field and then sent through a second field which took it into a detector *if its orientation was unchanged*. Between these two fields was placed an oscillating field whose frequency



could be varied. It was found that at certain very precise frequencies the number of atoms which reached the detector was considerably reduced, indicating that the frequency of alternation of the magnetic field was peculiarly efficient in causing nuclei to change orientation. Quite simple theory shows how this critical frequency is related to the magnetic field, the frequency of oscillation, and the magnetic moment of the nucleus. This is accordingly determined in terms of very accurately made measurements.

An ingenious modification of this idea was introduced by Purcell, Pound, and Torrey. They pointed out that if a considerable number of nuclei were changed in orientation there would need to be an absorption of energy, which could be measured. The energy derives from the potential energy of a magnetic moment in a magnetic field and is changed by reorientation of the moment. Purcell's modification consisted in using a whole block of nuclei, rather than a stream of molecules in a high vacuum, and observing the frequency at which energy absorption changes. The same relationship as in Rabi's method then applies, and a magnetic moment is determined.

With these methods the magnetic moments of proton, neutron, and deuteron have been measured. The moment of the deuteron is not the sum of the moments of its constituents. This is due to the fact that the deuteron spends part of its life history in an asymmetrical condition, and this in turn means that a part of the force between neutron and proton depends on the way in which the line joining the two elementary particles is directed with respect to the axes of the particles themselves.

### Mesons and nuclear forces

The outstanding theoretical difficulty in nuclear physics is the striking difference between the character of the force which is responsible for beta-ray decay (in which a neutron becomes a proton) which is quite weak, and the force which binds particles together in a nucleus which is very strong. This latter is, as we have said, at least partly due to an exchange process, and it would seem as though the exchange should be related to the conversion of a proton into a neutron and vice versa. In order to make the exchange process have more meaning Yukawa in 1935 postulated a particle of mass intermediate between that of a proton and an electron. This would account for the nature of the nuclear binding, in a qualitative way, even if it would not account for beta-ray decay forces. This particle he called a *meson*. The discovery of particles of this category in cosmic rays and the



production of such particles by nuclear bombardment as shown by Gardner and Lattes make it certain that Yukawa's suggestion has to be taken seriously.

The trouble arises in the mathematical and logical difficulties of setting up a theory of nuclear forces when none of our knowledge of the elementary particles is complete and where only the most advanced form of thought can be applied. Any theory has to be tentative in character, and the exhausting labor of mathematical manipulation and approximation does not appeal to a theorist unless he has compelling reasons for trying a hypothesis and excellent experimental data with which to test it. The needed data will almost certainly be forthcoming, as high-energy accelerators become commonplace and varied experimental genius is brought to bear on the problems.

### **High-energy physics: cosmic rays**

The subject of high-energy physics, in which sufficient energy is employed to shake the intimate structure of the nucleus, is just beginning. It occupies the same position with respect to the nuclear physics of simple transmutation and artificial radioactivity as the study of the nucleus has with respect to the electronic structure of the atom. Already one hears the title "classical nuclear physics" to refer to researches dealing with 10 Mev and lower energies. High-energy physics is a subject which is fascinating and exciting and combines operation of a large apparatus with the prospect of experimental results which are completely new and unexpected. To a scientist it is as adventurous as a trip to a star, and, indeed, in a sense that is what it is. The content of high-energy physics is found in nature in cosmic radiation, and the remarkable results of cosmic-ray research are now being dovetailed into research with huge cyclotrons in a manner which is rapidly showing what the content of nature at high energies is. Because of their great importance in this subject a word about cosmic rays is worth while.

Cosmic rays were discovered when excellent insulators and guarding techniques made it possible to measure very small amounts of ionization. At first their reality was under question, but experiments made in snow-fed lakes where no radioactivity could be expected in the water showed that there was indeed an ionizing radiation which could be absorbed by the water. The origin of this radiation is outside the earth, but its source is still not known today, forty years after its discovery.



Two reasons for this lack of understanding are properties of the earth itself. The earth has an atmosphere and also a magnetic field. In consequence we are never permitted to observe the true radiation coming in from somewhere out in space but have to put up with radiation which has been bent and filtered by the magnetic field and also has interacted many times with the atoms of the atmosphere. If in spite of these two complications we go ahead and study the nature of the penetrating radiation we find by far the strangest manifestation of matter. Cosmic rays seem to contain everything: electrons, positrons, photons, and neutrons, as well as other particles not yet sorted out. However, the most startling feature of cosmic rays is the existence of large releases of varied kinds of radiation, all at the same time and clearly having a common origin. These are called showers. These can be studied in two ways, either by an ionization chamber at very high pressure or by means of trays of Geiger counters and coincidence circuits. The first method reveals the existence of sudden bursts of ionization, amounting to as high as  $10^{15}$  electron volts. The second shows coincidences of many counters. If the counters are made to trigger a cloud chamber the chamber is seen to be filled with a tremendous confusion of ionizing particles having apparently a common origin. A magnetic field applied across the cloud chamber shows that the particles are of either sign. In fact the discovery of the positron was made by Anderson in the study of cosmic rays and not by Curie and Joliot in the study of artificial radioactivity.

The fact that the particles in these showers have a common origin means that there must be single particles entering the atmosphere and having very large energies. These particles are called the primary radiation. They seem to arrive from all directions though some slight directivity has been reported. It seems hard to believe that the process of nuclear release of energy going on in stars could give rise to such enormous energies concentrated in single particles. Therefore, thought is turning to possible methods of acceleration. This is again a wide-open subject since little is known about interstellar fields. However, if the evidence from the doppler shift of the nebulae is right, then the universe has had about  $3 \times 10^9$  years for acceleration processes to take place, and this is long enough for quite slow accelerations to produce an effect. Whatever reason the reader likes to imagine for this acceleration, there seems to be no doubt that very energetic charged particles are entering the earth's domain and that these are responsible for cosmic radiation.



The use of photographic plates sent up to great altitudes by means of balloons has shed considerable light on the nature of the heavy particles reaching the earth. The development of the plates reveals the presence of densely ionizing tracks, of the kind shown in Fig. 3. These tracks have spurs called "delta rays," and the frequency of these along the track is sharply dependent on the charge carried by the incoming

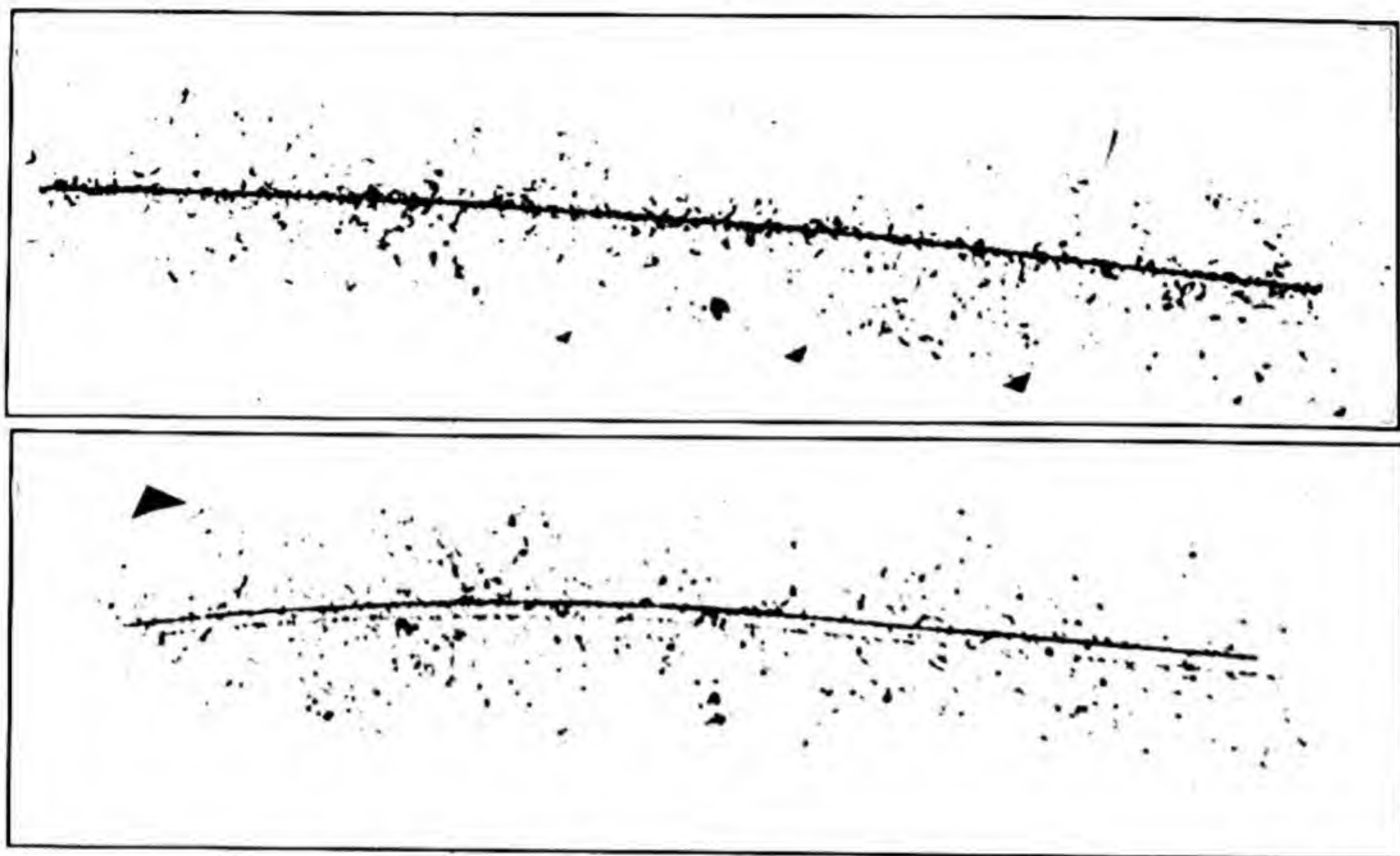


FIG. 3. Microphotograph of the track of a heavy primary of charge  $Z = 24 \pm 2$  as it appears in three plates before this nucleus suffers a collision in the glass between the third and the fourth emulsion. The lower picture is a microphotograph of the track of the heaviest fragment of charge  $Z = 11 \pm 1$  resulting from the collision, as it appears in the fourth emulsion, accompanied by an  $\alpha$ -particle "satellite track" and six proton tracks. The apparent curvature of the track results from a distortion of the emulsion. These pictures are due to Dr. H. Bradt.

particle. The greater this charge, the more and longer the delta rays. By making a count of the frequency of the delta rays, the charge of the particle can be found, and the discovery has been made that the agents causing the tracks are stripped nuclei of elements up to iron. The cosmic rays therefore represent bombardment of the earth by atomic nuclei and in turn give us some idea of the abundance of elements in the universe. Hydrogen seems to be very abundant, and the suggestion that protons form a large part of the primary radiation was first made by Johnson. The extension to include elements up to number 26 was first made by Freier, Lofgren, Ney, F. Oppenheimer, Bradt, and Peters on the basis of the photographic work just described.



With this discovery one would expect that the known processes of interaction with magnetic fields on the one hand and of the atmosphere on the other would render the understanding of cosmic rays easy. To some extent this is true. For example there is no tremendous problem posed by a shower, providing the primary particle is capable of interacting with matter to produce radiation and electrons, for then the radiation will produce electron positron pairs, which in turn will produce more radiation, and so on, the whole complex process being due to the conversion of the kinetic energy of the incoming particle into the kinetic energy of many particles. A man-made shower produced by high-energy photons from a synchrotron is shown in Fig. 4. It has actually proved, however, that the interaction of the primary particle with matter is not only to produce electrons as ordinarily known. This was suspected first when it was realized that an electron when going at high speed will produce radiation if it is being decelerated by matter. This radiation is identical with the continuous x-radiation produced in an x-ray tube but is much more prolific for high-energy electrons. The electron loses energy fast. Therefore no such electrons should be capable of penetrating the atmosphere. This statement is not borne out by experience because penetrating particles do exist.

The explanation for this discrepancy has been found in the existence of *heavy* electrons, called mesons. These are in some way concerned with the nucleus, in accordance with Yukawa's suggestion. Because they are heavier than electrons they radiate much less, and so although they ionize more they are actually on the whole more penetrating. The discovery of the meson is of some interest especially because it has been found that there are at least two kinds of meson. There exist two ways of deciding the mass of a particle: one can measure its total energy and one can measure its velocity separately. Its mass can then be found. Or one can measure its momentum and then its velocity, so finding the mass. The latter method was used by Anderson and Neddermyer at the California Institute of Technology and Street and Stevenson at Harvard. The momentum of a particle is measurable by its curvature in a magnetic field. On the other hand the velocity can be measured by the rate of ionization, which is determined by the electric field of the particle and how fast it goes by, and so depends only on the charge and the velocity. In point of fact, a meson of kinetic energy  $T$  and mass  $M$  (electron masses) loses energy by ionization like an electron of kinetic energy  $T/M$ . Therefore if the curvature in a magnetic field can be measured and also the rate of



formation of ions it is possible to determine the mass of the meson. The value found by this method is 198 electron masses.

The first method was employed by Powell and Occhialini, who employed the photographic technique. This is very convenient because



FIG. 4. A man-made shower produced by the gamma radiation produced by bombardment of a target by 335-Mev electrons from the University of California synchrotron. This remarkable picture shows the process of interchange between matter and radiation which constitutes a shower. The gamma radiation enters at the left, and a fraction is converted into particle energy in the first lead plate. Some of this particle energy is reconverted into lower energy radiation in the second lead plate. The original single quantum is now multiple, being partly particle energy and partly lower energy quanta. About the middle there is a maximum of energy in ionizing particles, and thereafter the lead plates succeed in absorbing the energy. (Radiation Laboratory of the University of California, and the Atomic Energy Commission.)

by counting the grains along the track of a cosmic ray particle which stops in the emulsion it is possible to estimate its total energy. By counting the grains at various stages of its path it is possible to compare its rate of ionization with known particles such as protons or electrons. Powell also found that there were mesons in cosmic



rays, but he found several cases of a double process, which is illustrated in Fig. 5. Here a particle which is undoubtedly neither proton nor electron is seen to proceed along its career, developing more ability to ionize as it goes, and then at the end of its path a fresh particle, also a meson, is seen to appear, which follows a career to a conclusion. By careful measurements it can be shown that the first particle is heavier than the second, having 286 electron masses. Powell called it a  $\pi$ -meson, and the lighter form a  $\mu$ -meson. The  $\mu$ -meson is the same as that of Anderson and Street.

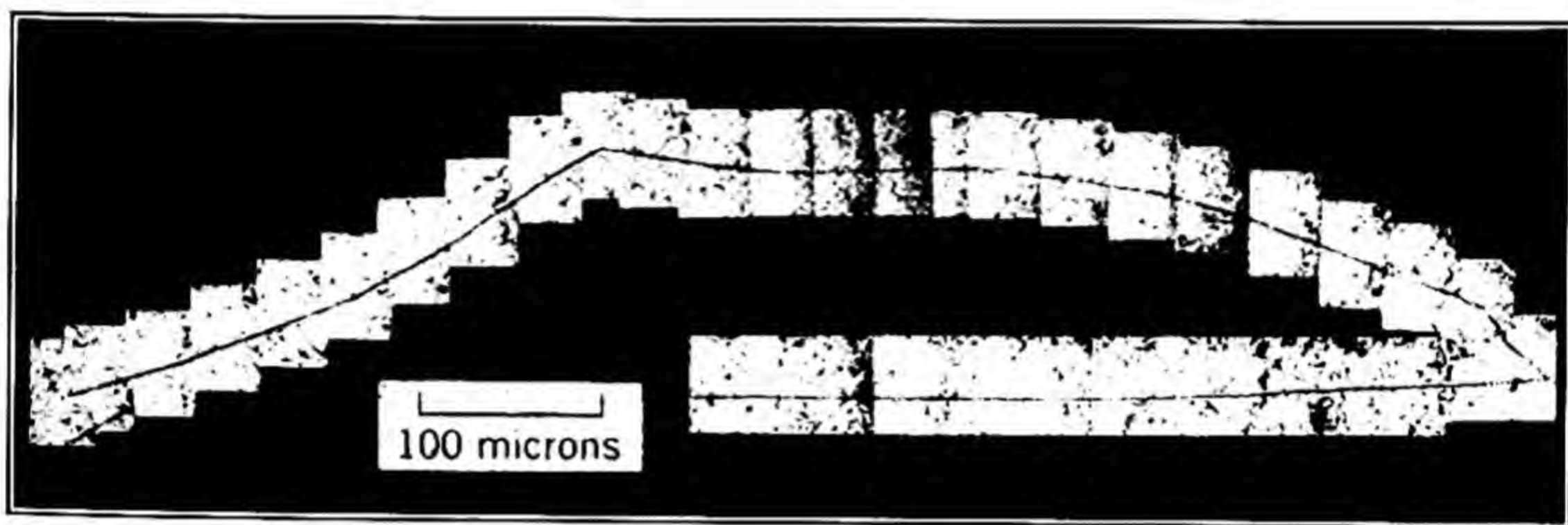


FIG. 5. A photograph of the life history of a  $\pi$ -meson, due to the joint program of the Atomic Energy Commission and the Radiation Laboratory of the University of California. The  $\pi$ -meson enters from the extreme left in the mosaic of pictures and converts into a  $\mu$ -meson at the top right. This meson, whose lower mass is apparent in its more ready scattering, moves from left to right in the upper mosaic until at the sharp break it is converted into an electron and a neutrino. The electron has high energy and ionizes very sparsely. It leaves the picture at the left of the lower mosaic.

This heavier  $\pi$ -meson has been observed to be produced by fast alpha particles, protons, or electrons in cyclotrons and synchrotrons, and it now seems clear that only the  $\pi$ -meson plays any active part in the nucleus.

The meson story therefore appears to be as follows: There exist positive and negative  $\pi$ -mesons. Neither are permanent, nor do they have similar histories. The negative  $\pi$ -meson is actually strongly attracted to nuclei. The cross section for a transmutation by a negative  $\pi$ -meson is therefore very large, and the kind of transmutation it produces is quite an event since it carries with it 150 Mev of mass energy apart from its kinetic energy. Any nucleus foolish enough to attract into its midst such a character is immediately blown apart into fragments and these are recognizable as "stars," one of which is shown in Fig. 12, Chapter 5. The free half-life of such a meson has recently



been measured by Richardson who has found that the rate of production of stars in photographic emulsion drops off if the emulsion is placed further and further along the path of a negative  $\pi$ -meson. No collisions occur in the high vacuum of a cyclotron, so this seemingly represents the decay of the  $\pi$ -meson to a  $\mu$ -meson. The half-life is  $8 \times 10^{-9}$  second.

The positive  $\pi$ -meson is different. It is not welcome in nuclei, though it can force its way in and then produce a star. It appears to be able to come to rest and then change into a  $\mu$ -meson by the process shown in Fig. 5. It seems likely that the process of  $\pi \rightarrow \mu$  decay takes the same time for either positive or negative  $\pi$ -mesons.

$\mu$ -mesons predominantly decay into a fast electron and a neutrino as seen in Fig. 5. This decay process has been observed in two ways. The first is by noting the loss of  $\mu$ -mesons in going through sparsely absorbing matter (and hence taking time as well as losing energy) to be greater than in dense matter (where energy is lost without waste of time). The second is the ingenious method, due to Rasetti, of letting the meson record in a counter, then plough to a stop in a block of lead, and detecting the emitted fast electron after decay. The fast electron is not in coincidence with the meson count *unless the meson count is delayed*. Measurement of this delay by a circuit measures the half-life. As measured by the first method the half-life is  $2 \times 10^{-4}$  second. Correcting for the fact that the timing system is slowed down by the motion of the particle so that time is lengthened to be  $1/\sqrt{1 - (v^2/c^2)}$  too long, the half-life proves to be  $2 \times 10^{-6}$  second, which agrees with the measurements made by the second method where the meson is at rest with respect to the observer. A negative  $\mu$ -meson can also be captured by nuclei in a manner corresponding to a rather weak force. After capture they apparently give rise to neutrons and neutrinos.

In addition neutral mesons may be present and the processes may also involve a neutrino. It is likely that  $\mu$ -mesons have a spin of  $\frac{1}{2}$  whereas  $\pi$ -mesons have a spin of 1.

There seem to be still heavier mesons. At the time of writing these have been found only in cosmic rays and study of them is slow and tedious. Full knowledge of mesons is far from being realized.

To return to nuclear forces, the  $\pi$ -meson has not yet produced the surge forward in theory that might be expected. Several meson theories of nuclear forces exist, but so far they give no simple clarification. Part of this is because the properties of the  $\pi$ -meson are not com-



pletely known and part because the theory is abstract and not adequately guided by experiment. Very accurate scattering work seems to be needed, and this takes time. The large nuclear machines with pulsed beams and fast particles are by no means so easy to use for experiments as they might seem. For this reason the twist in thought that will bring the nucleus within the scope of simple ideas has not yet been found.

To leave on this rather unsatisfactory note gives us genuine pleasure. This is an age of vanishing frontiers, and it adds a touch of vigor to consider that here the frontier is very much present. We doubt whether the complete understanding of the atomic nucleus will be attained in our lifetime, and this adds considerable zest to existence. The feeling of zest is shared by workers in lab coats awaiting the 1000-meter beam of the supercyclotron, by workers patiently amassing data about cosmic rays, by workers operating calculating machines and wearing down pencils. A little of it should have reached the reader.

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- H. A. BETHE, *Elementary Nuclear Theory*, John Wiley and Sons, Inc., 1946.  
*Cosmic Radiation*, a symposium, Interscience Publishers, New York, 1949.



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## APPENDIX 1

### DATES OF SOME IMPORTANT DEVELOPMENTS

- Discovery of alpha particle, *Rutherford*, 1904.  
Scintillations, *Crookes*, and *Elster and Geitel*, 1903.  
Cloud chamber used for nuclear-particle detection, *Wilson*, 1912.  
Nuclear atom, *Rutherford and Bohr*, 1912.  
Continuous beta-ray spectrum, *Chadwick*, 1914.  
*Rutherford-Geiger* counter, 1908.  
Isotopes of neon, *Thomson*, 1912.  
Thermal diffusion predicted, *Chapman, Borelius*, 1915.  
Transmutation of nitrogen, *Rutherford*, 1919.  
Further transmutations by *Rutherford and Chadwick*, 1919-1923.  
Mass spectrograph, *Aston*, 1919.  
Radioactive indicators used, *von Hevesy and Paneth*, 1913.  
Radio-autographs, *Lacassagne and Lattes*, 1924.  
Physiological effects of x- and gamma rays discovered, 1898.  
Genetic effect of x-rays, *Muller*, 1927.  
Oil diffusion pumps, 1928.  
Large water-cooled triodes, 1926.  
*Geiger-Müller* counter, 1928.  
Cyclotron first described, *Lawrence*, 1932.  
Transmutation of lithium by artificially accelerated protons, *Cockcroft and Walton*, 1932.  
Deuterium discovered, *Urey, Brickwedde, and Murphy*, 1932.  
*Van de Graaff* machine, 1932.  
Neutron discovered, *Chadwick*, 1932.  
Positron discovered, *Anderson*, 1932.  
Artificial radioactivity discovered, *Curie and Joliot*, 1934.  
Cyclotron shimming discovered, 1934.  
Large-scale separation of neon isotopes, *Hertz*, 1934.  
Rubber gasket vacuum seals, 1936.  
Discovery of  $\text{Na}^{24}$ ,  $\text{P}^{32}$ , 1935.  
Discovery of  $\text{Fe}^{59}$ , 1937.  
Discovery of  $\text{I}^{131}$ , 1939.  
Theoretical limit to cyclotron energy set at 10 Mev, 1938.  
Crocker Laboratory beam found, 16 Mev, 1939.  
In 1932, no artificial radioactive elements.  
In 1934, 3 artificial radioactive elements.  
In 1937, 190 artificial radioactive isotopes.  
In 1939, 270 artificial radioactive isotopes.  
In 1941, 370 artificial radioactive isotopes.



- In 1949, 800 artificial radioactive isotopes.  
Uranium fission discovered, *Hahn and Strassmann*, 1938.  
Chemical exchange method of isotope separation, *Urey*, 1936.  
Thermal diffusion method of isotope separation, *Clusius and Dickel*, 1937.  
Leukemia therapy tried, *J. Lawrence*, 1938.  
Metabolic isotope lability discovered, *Schoenheimer*, 1939.  
Nuclear emulsions developed, *Blau*, 1924; *Wilkins*, 1937.  
Discovery of  $C^{14}$ , *Ruben and Kamen*, 1940.  
Betatron developed, *Kerst*, 1941.  
 $\mu$ -meson discovered, *Anderson and Street*, 1939.  
First self-sustained nuclear chain reaction, Chicago, 1942.  
Electromagnetic isotope separation in quantity, 1942.  
Diffusion method of isotope separation, 1942.  
Large-scale production of plutonium, 1945.  
First nuclear explosion, 1945.  
Synchrotron principle discovered, *McMillen, Veksler*, 1945.  
Neutron diffraction tried, 1945.  
Photomultiplier scintillation counter, *Kallmann*, 1946.  
 $\pi$ -meson discovered, *Powell*, 1946.  
Frequency-modulated cyclotron operated, *Richardson*, 1946.  
Synchrotron operated, *London*, 1946.  
Radioactive isotopes from the pile available, 1946.  
184-in. Berkeley cyclotron operated, 1947.  
Artificial production of  $\pi$ -mesons, *Gardner and Lattes*, 1948.



## APPENDIX 2

### TABLE OF ATOMIC SPECIES

In this section of the Appendix we give a table of atomic species. Of these, many are stable, many are radioactive with the emission of electrons, and many are positron emitters. These three categories almost cover the whole range of possibilities, and so we have attempted to make the table as compact as possible by indicating which of the three applies to a certain element, as follows. Stable elements are listed in **bold-face** type. If they are naturally occurring but radioactive, this fact is stated. Beta-ray emitters are listed in ordinary type, and positron emitters appear in *italics*. The second column of the table shows either the abundance or the half-life, according to whether the element is stable or radioactive. The last two columns show the energies of the radiations. Only if the categorical statement appears should it be assumed that a blank space means the absence of a radiation; it generally means that the information has not yet been obtained experimentally.

The authors have had much difficulty in deciding what should be the aim in compiling this table. The entire list is necessarily incomplete because the ability to create and study artificial isotopes is increasing enormously, first, on account of nuclear fission, and, more important, on account of high-energy bombardment which has tremendous power for nuclear synthesis followed by nuclear adaptation. We therefore recommend for complete information the article by Seaborg and Perlman in *Reviews of Modern Physics*, October 1948. We wish to add our tribute to the service rendered by their compilation of data on radioactivity. Anyone intending to do serious research in artificial radioactivity should keep his own isotope table up to date with the aid of the current literature.

Our decision has been to include all readily accessible radioactive isotopes but to be sketchy on those produced in fission, whether by slow neutrons or energetic bombardment. We call attention to the considerable extent of gamma-ray decay, otherwise known as nuclear isomerism, and many such cases can be recognized in the table by  $\gamma$ . Alpha decay is indicated by  $\alpha$ . Under the "particle energies" column is occasionally an electron released from inside the atomic shells but correlated with gamma radiation, the process of internal conversion. The notation ( — ) indicates this fact.

For many elements the actual decay scheme is quite complex. Original sources, which are admirably given in Seaborg and Perlman's article, should be consulted for these.

Not many general rules can be given about atomic species. One simple generality holds, namely, that practically no elements of odd atomic number and even mass number are stable. In addition, elements of odd atomic number are



found with either one isotope only, or two of nearly equal abundance. The large numbers of isotopes are found with even atomic number. An interesting question concerns *isobars*, isotopes with different atomic numbers but the same mass number. Many such examples are known, but most of them differ in atomic number by more than one unit. This indicates that the change of one isobar into the other requires transition through a third isobar, and there may be insufficient energy available for the change. If, however, isobars existed with atomic number differing by one unit, one would presumably be more stable than the other, and the question arises why the transition does not take place. Actually several such isobars are known, for example,  $\text{In}^{113}$  and  $\text{Cd}^{113}$ , and both are stable. This fact is proof of the existence of strong selection rules which may greatly modify, or even prevent, an otherwise expected transition. In other words, one or the other of these pairs of isobars is radioactive, but with a nearly infinite half-life.

Even though this is a rather dull and long table we cannot resist a short comment on its significance. This table is the code which contains the history of the earth. The present abundance of the elements of long half-life tell us its age, around  $3 \times 10^9$  years. The relative frequency of occurrence of various isotopes distributed throughout tell us how elements were put together. This we do not yet know, though just recently an attractive hypothesis in terms of neutron capture has been put forth. Study of this dull table, after knowledge of nuclear theory has advanced, will influence cosmology, biology, and anthropology. Its compilation is therefore of no mean importance.

ATOMIC NUMBER	ATOMIC SPECIES	ABUNDANCE OR HALF-LIFE	PARTICLE ENERGIES (Mev)	QUANTUM ENERGIES
0	$n^1$	15 m *	0.8	
1	$\text{H}^1$	99.98%		
	$\text{H}^2$	0.02%		
	$\text{H}^3$	12.4 y *	0.019	
2	$\text{He}^3$	$10^{-5}\%$		
	$\text{He}^4$	100%		
		Mass five missing		
	$\text{He}^6$	0.8 s *	3.7	
3	$\text{Li}^6$	7.9%		
	$\text{Li}^7$	92.1%		
	$\text{Li}^8$	0.88 s	12.7 followed by alpha decay	
4	$\text{Be}^7$	K		0.45
	$\text{Be}^9$	100%		
	$\text{Be}^{10}$	$2.7 \times 10^6$ y	0.56	
5	$\text{B}^{10}$	18.8%		
	$\text{B}^{11}$	31.2%		
	$\text{B}^{12}$	0.022 s	13.43	

\* y = year(s); s = second(s); m = minute(s); h = hour(s); d = day(s).



ATOMIC NUMBER	ATOMIC SPECIES	ABUNDANCE OR HALF-LIFE	PARTICLE ENERGIES (Mev)	QUANTUM ENERGIES
6	$C^{10}$	19.1 s	2.2	1.0
	$C^{11}$	21.0 m	0.95	
	$C^{12}$	98.9%		
	$C^{13}$	1.1%		
	$C^{14}$	5000 y	0.145	
7	$N^{12}$	0.012 s	16.6	6.7, 6.2
	$N^{13}$	10.1 m	1.24	
	$N^{14}$	99.62%		
	$N^{15}$	0.38%		
	$N^{16}$	7.35 s	10.5, 4.6, 4.3, 3.8	
	$N^{17}$	4.1 s	3.7, also neutrons	
8	$O^{14}$	76 s	1.8	2.3
	$O^{15}$	126 s	1.7	
	$O^{16}$	99.76%		
	$O^{17}$	0.04%		
	$O^{18}$	0.20%		
	$O^{19}$	27 s	4.5, 2.9	
9	$F^{17}$	70 s	2.1	2.2
	$F^{18}$	112 m	0.7	
	$F^{19}$	100%		
	$F^{20}$	12 s	5.0	
10	$Ne^{19}$	20.3 s	2.20	
	$Ne^{20}$	90.0%		
	$Ne^{21}$	0.27%		
	$Ne^{22}$	9.73%		
	$Ne^{23}$	40 s	4.1	
11	$Na^{21}$	23 s		1.3
	$Na^{22}$	3.0 y	0.58	
	$Na^{23}$	100%		
	$Na^{24}$	14.8 h	1.4	
	$Na^{25}$	58 s	3.7, 2.7	
12	$Mg^{23}$	11.6 s	2.84	1.8, 1.3
	$Mg^{24}$	77.4%		
	$Mg^{25}$	11.5%		
	$Mg^{26}$	11.1%		
	$Mg^{27}$	10.2 m		
13	$Al^{25}$	8 s		1.9
	$Al^{26}$	7 s	2.99	
	$Al^{27}$	100%		
	$Al^{28}$	2.4 m	2.75	
	$Al^{29}$	6.7 m	2.5	
14	$Si^{27}$	5 s	3.7	
	$Si^{28}$	92.3%		



ATOMIC NUMBER	ATOMIC SPECIES	ABUNDANCE OR HALF-LIFE	PARTICLE ENERGIES (Mev)	QUANTUM ENERGIES
15	Si <sup>29</sup>	4.7%		
	Si <sup>30</sup>	3.0%		
	Si <sup>31</sup>	170 m	1.8	None
	P <sup>29</sup>	4.6 s	3.6	
	P <sup>30</sup>	2.55 m	3.0	
	P <sup>31</sup>	100%		
16	P <sup>32</sup>	14.30 d *	1.71	None
	S <sup>31</sup>	2.6 s		
	S <sup>32</sup>	95.0%		
	S <sup>33</sup>	0.74%	2	
	S <sup>34</sup>	4.2%		
	S <sup>35</sup>	87 d	0.17	
	S <sup>36</sup>	0.016%		
	S <sup>37</sup>	5.0 m	4.3, 1.6	2.6
17	Cl <sup>33</sup>	2.8 s		
	Cl <sup>34</sup>	33 m		
	Cl <sup>35</sup>	75.4%		
	Cl <sup>36</sup>	10 <sup>6</sup> y	0.66, also K and positron	
	Cl <sup>37</sup>	24.6%		
	Cl <sup>38</sup>	37 m	1.1, 3.2, 5.0	1.7, 2.0
	Cl <sup>39</sup>	1 h *		
	A <sup>35</sup>	1.9 s	4.4	
	A <sup>36</sup>	0.307%		
18	A <sup>37</sup>	34.1 d	K	None
	A <sup>38</sup>	0.061%		
	A <sup>39</sup>	2.5 m		
	A <sup>40</sup>	99.632%		
	A <sup>41</sup>	110 m	1.2	1.37
	K <sup>38</sup>	7.5 m	2.3	
	K <sup>39</sup>	93.3%		
19	K <sup>40</sup>	0.012%	Naturally radio- active, 1.4 elec- tron, 0.5 positron, K	1.5
	K <sup>41</sup>	6.7%		
	K <sup>42</sup>	12.4 h	3.6, 2.1	1.5
	Ca <sup>40</sup>	96.96%		
	Ca <sup>42</sup>	0.64%		
20	Ca <sup>43</sup>	0.15%		
	Ca <sup>44</sup>	2.06%		
	Ca <sup>45</sup>	180 d	0.25	None
	Ca <sup>46</sup>	0.0033%		
	Ca <sup>47</sup>	5.8 d	1.1	1.3



ATOMIC NUMBER	ATOMIC SPECIES	ABUNDANCE OR HALF-LIFE	PARTICLE ENERGIES (Mev)	QUANTUM ENERGIES
21	$\text{Ca}^{48}$	<b>0.19%</b>		
	$\text{Ca}^{49}$	2.5 h	2.3	0.8
	$\text{Sc}^{41}$	0.8 s	4.9	
	$\text{Sc}^{42}$	13.5 d	1.4	
	$\text{Sc}^{43}$	4 h	0.4, 1.4	1.0
	$\text{Sc}^{44}$	52 h	Isomer	0.26
	$\text{Sc}^{44}$	4.1 h	1.5	
	$\text{Sc}^{45}$	<b>100%</b>		
	$\text{Sc}^{46}$	85 d	0.36, K	1.1, 0.9
	$\text{Sc}^{48}$	44 h	0.5, 1.4	0.9
	$\text{Sc}^{49}$	57 m	1.8	None
22	$\text{Ti}^{45}$	3.0 h	1.2	
	$\text{Ti}^{46}$	<b>7.95%</b>		
	$\text{Ti}^{47}$	<b>7.75%</b>		
	$\text{Ti}^{48}$	<b>73.45%</b>		
	$\text{Ti}^{49}$	<b>5.51%</b>		
	$\text{Ti}^{50}$	<b>5.34%</b>		
	$\text{Ti}^{51}$	6 m	Isomer, 1.6	
	$\text{Ti}^{51}$	72 d	0.36	1.0
23	$\text{V}^{47}$	600 d	K	None
	$\text{V}^{48}$	16 d	1.0	1.05
	$\text{V}^{49}$	33 m	1.9	
	$\text{V}^{50}$	3.7 h		
	$\text{V}^{51}$	<b>100%</b>		
	$\text{V}^{52}$	3.9 m	2.05	
24	$\text{Cr}^{49}$	45 m		
	$\text{Cr}^{50}$	<b>4.49%</b>		
	$\text{Cr}^{51}$	26.5 d	Also K	0.32
	$\text{Cr}^{52}$	<b>83.77%</b>		
	$\text{Cr}^{53}$	<b>9.43%</b>		
	$\text{Cr}^{54}$	<b>2.30%</b>		
	$\text{Cr}^{55}$	2 h		
25	$\text{Mn}^{51}$	46 m	2.0	
	$\text{Mn}^{52}$	21 m	0.6, K	1.4, 0.9, 0.7, 0.4
	$\text{Mn}^{53}$	6.5 d	0.77	1.0
	$\text{Mn}^{54}$	310 d	K	0.85
	$\text{Mn}^{55}$	<b>100%</b>		
	$\text{Mn}^{56}$	2.59 h	2.8, 1.0, 0.7	0.8, 1.8, 2.1
26	$\text{Fe}^{53}$	8.9 m		
	$\text{Fe}^{54}$	<b>6.04%</b>		
	$\text{Fe}^{55}$	4 y	K	
	$\text{Fe}^{56}$	<b>91.57%</b>		
	$\text{Fe}^{57}$	<b>2.11%</b>		
	$\text{Fe}^{58}$	<b>0.28%</b>		



ATOMIC NUMBER	ATOMIC SPECIES	ABUNDANCE OR HALF-LIFE	PARTICLE ENERGIES (Mev)	QUANTUM ENERGIES
	Fe <sup>59</sup>	47 d	0.26, 0.46	1.1, 1.3
27	Co <sup>55</sup>	18.2 h	1.50	0.16, 0.21, 0.8, 1.2
	Co <sup>56</sup>	72 d	1.2	1.05
	Co <sup>57</sup>	0.17%		
	Co <sup>57</sup>	270 d	0.26	
	Co <sup>58</sup>	72 d	0.4	0.6
	Co <sup>59</sup>	99.83%		
	Co <sup>60</sup>	5.3 y	0.31	1.16, 1.32
	Co <sup>60</sup>	10.7 m		
	Co <sup>61</sup>	1.75 h	1.1	None
28	Ni <sup>57</sup>	2 m		
	Ni <sup>58</sup>	68.0%		
	Ni <sup>59</sup>	36 h	0.67	
	Ni <sup>60</sup>	27.2%		
	Ni <sup>61</sup>	0.1%		
	Ni <sup>62</sup>	3.8%		
	Ni <sup>63</sup>	2.6 h	1.9	1.1
	Ni <sup>64</sup>	0.9%		
	Ni <sup>65</sup>	2.6 h		
29	Cu <sup>58</sup>	10 m		
	Cu <sup>59</sup>	81 s		
	Cu <sup>60</sup>	25 m	1.8, 3.3	1.5
	Cu <sup>61</sup>	34 h	0.9	None
	Cu <sup>62</sup>	10.5 m	2.6	
	Cu <sup>63</sup>	68%		
	Cu <sup>64</sup>	12.8 h	0.58, also posi- tron, 0.66, K	Weak 1.35
	Cu <sup>65</sup>	32%		
	Cu <sup>66</sup>	5 m	2.6	1.3
30	Zn <sup>63</sup>	38 m	2.3	Weak 1.0, 1.9
	Zn <sup>64</sup>	50.9%		
	Zn <sup>65</sup>	250 d	0.4	0.45, 0.65, 1.1
	Zn <sup>66</sup>	27.3%		
	Zn <sup>67</sup>	3.9%		
	Zn <sup>68</sup>	17.4%		
	Zn <sup>69</sup>	13.8 h	Isomer	0.47
	Zn <sup>69</sup>	57 m	1.0	None
	Zn <sup>70</sup>	0.5%		
31	Ga <sup>64</sup>	48 m		
	Ga <sup>65</sup>	15 m	K	0.054, 0.117
	Ga <sup>66</sup>	9.4 h	3.1	
	Ga <sup>67</sup>	83 h	K	0.18, 0.30
	Ga <sup>68</sup>	68 m	1.9	
	Ga <sup>69</sup>	60.2%		
	Ga <sup>70</sup>	20 m	1.7	



ATOMIC NUMBER	ATOMIC SPECIES	ABUNDANCE OR HALF-LIFE	PARTICLE ENERGIES (Mev)	QUANTUM ENERGIES
32	<b>Ga<sup>71</sup></b>	<b>39.8%</b>		
	Ga <sup>72</sup>	14 h	2.4, 0.8	2.4, 0.8, 0.6
	Ga <sup>73</sup>	5 h	1.4	None
	Ge <sup>66</sup>	140 m		
	Ge <sup>67</sup>	23 m		
	Ge <sup>68</sup>	250 d	K	
	<b>Ge<sup>70</sup></b>	<b>21.2%</b>		
	Ge <sup>71</sup>	11 d	Isomer	
	Ge <sup>71</sup>	40 h	1.2	
	<b>Ge<sup>72</sup></b>	<b>27.3%</b>		
	<b>Ge<sup>73</sup></b>	<b>7.9%</b>		
	<b>Ge<sup>74</sup></b>	<b>37.1%</b>		
	Ge <sup>75</sup>	89 m	1.1	
	<b>Ge<sup>76</sup></b>	<b>6.5%</b>		
33	Ge <sup>77</sup>	12 h	1.9	
	Ge <sup>77</sup>	59 s	2.8	
	As <sup>71</sup>	52 m		
	As <sup>71</sup>	60 h	K	
	As <sup>72</sup>	26 h	2.8	2.4
	As <sup>73</sup>	90 d	K	
	As <sup>74</sup>	18 d	1.3(−) 0.9(+)	0.58
	<b>As<sup>75</sup></b>	<b>100%</b>		
34	As <sup>77</sup>	40 h	0.8	
	As <sup>78</sup>	80 m	1.4	0.27
	Se <sup>71</sup>	44 m		
	Se <sup>72</sup>	9.5 d	K	
	Se <sup>73</sup>	7.0 h	1.29	
	<b>Se<sup>74</sup></b>	<b>0.87%</b>		
	Se <sup>75</sup>	123 d	K	0.40, 0.28, 0.13
	<b>Se<sup>76</sup></b>	<b>9.02%</b>		
	<b>Se<sup>77</sup></b>	<b>7.58%</b>		
	Se <sup>77</sup>	17.5 s	0.13	0.15
	<b>Se<sup>78</sup></b>	<b>23.52%</b>		
	<b>Se<sup>80</sup></b>	<b>49.82%</b>		
	Se <sup>81</sup>	59 m		
	Se <sup>81</sup>	18 m	1.5	None
	<b>Se<sup>82</sup></b>	<b>9.19%</b>		
	Se <sup>83</sup>	67 s	3.4	
	Se <sup>83</sup>	27 m	1.5	1.1, 0.37, 1.1
	Se <sup>84</sup>	4 m		
35	Br <sup>75</sup>	1.7 h	1.6, K	None
	Br <sup>76</sup>	15.7 h	3.15, also 0.18(−)	2
	Br <sup>77</sup>	57 h	0.36	0.7
	Br <sup>78</sup>	6.4 m	2.3	0.1, 0.05
	<b>Br<sup>79</sup></b>	<b>50.5%</b>		



ATOMIC NUMBER	ATOMIC SPECIES	ABUNDANCE OR HALF-LIFE	PARTICLE ENERGIES (MeV)	QUANTUM ENERGIES
36	Br <sup>80</sup>	4.4 h		
	Br <sup>80</sup>	18 m	2.0( - ) 0.73( + )	0.4
	Br <sup>81</sup>	49.5%		
	Br <sup>82</sup>	34 h	0.47	1.35, 0.79, 0.55
	Br <sup>83</sup>	2.4 h	1.0	None
	Br <sup>84</sup>	30 m	5.0	
	Br <sup>85</sup>	3.0 m	2.5	None
	Br <sup>87</sup>	55 s	0.25, followed by neutron	
	Br <sup>87</sup>	4.5 s	0.43, followed by neutron	
	Br <sup>88</sup>	16 s		
	Kr <sup>77</sup>	1.1 h	1.7, K	
	Kr <sup>78</sup>	0.342%		
	Kr <sup>79</sup>	3.4 h	0.9, 0.6, 0.4, K, 0.2	
	Kr <sup>80</sup>	2.22%		
	Kr <sup>82</sup>	11.50%		
	Kr <sup>83</sup>	11.48%		
	Kr <sup>83</sup>	113 m		
	Kr <sup>84</sup>	57.0%		
	Kr <sup>85</sup>	9.4 y	0.74	None
	Kr <sup>86</sup>	17.43%		
	Kr <sup>87</sup>	74 m	4	
	Kr <sup>88</sup>	3 h	2.5	
	Kr <sup>89</sup>	2.6 m		
	Kr <sup>90</sup>	33 s		
	Kr <sup>91</sup>	7 s		
	Kr <sup>92</sup>	2.3 s		
	Kr <sup>93</sup>	2.2 s		
	Kr <sup>94</sup>	1.4 s		
37	Rb <sup>81</sup>	5.0 h	0.9, 0.2( - )	0.8
	Rb <sup>82</sup>	6.4 h	0.9	1.0
	Rb <sup>84</sup>	40 d		
	Rb <sup>85</sup>	72.8%		
	Rb <sup>86</sup>	19.5 d	1.8, 0.7	1.08
	Rb <sup>87</sup>	27.2% (6 × 10 <sup>10</sup> y, naturally active)	0.13	
	Rb <sup>88</sup>	17.5 m	4.9	
	Rb <sup>89</sup>	15 m	3.8	
38	Sr <sup>84</sup>	0.56%		
	Sr <sup>85</sup>	70 m		0.17
	Sr <sup>85</sup>	15 d	K	0.8
	Sr <sup>86</sup>	3.86%		
	Sr <sup>87</sup>	2.7 h		0.37



ATOMIC NUMBER	ATOMIC SPECIES	ABUNDANCE OR HALF-LIFE	PARTICLE ENERGIES (Mev)	QUANTUM ENERGIES
39	$\text{Sr}^{87}$	<b>7.02%</b>		
	$\text{Sr}^{88}$	<b>82.56%</b>		
	$\text{Sr}^{89}$	54 d	1.5	None
	$\text{Sr}^{90}$	25 y	0.6	None
	$\text{Sr}^{91}$	10 h	3.2, 1.3	1.3
	$\text{Sr}^{92}$	2.7 h		
	$\text{Y}^{87}$	14 h		0.5
	$\text{Y}^{87}$	80 h	<i>K</i>	
	$\text{Y}^{88}$	2.0 h	1.5	
	$\text{Y}^{88}$	105 d	0.8	2.7, 1.8, 0.9
	$\text{Y}^{89}$	<b>100%</b>		
	$\text{Y}^{90}$	62 h	2.3	None
	$\text{Y}^{91}$	51 m		0.61
	$\text{Y}^{91}$	57 d	1.5	None
40	$\text{Y}^{92}$	3.5 h	3.5	1.0
	$\text{Y}^{93}$	10.5 h	3.1	0.7
	$\text{Y}^{94}$	20 m		
	$\text{Zr}^{89}$	80 h	1.1	None
	$\text{Zr}^{90}$	<b>51.46%</b>		
	$\text{Zr}^{91}$	<b>11.23%</b>		
	$\text{Zr}^{92}$	<b>17.11%</b>		
	$\text{Zr}^{94}$	<b>17.40%</b>		
	$\text{Zr}^{95}$	65 d	1.0, 0.4	0.9, 0.7, 0.2
41	$\text{Zr}^{96}$	<b>2.80%</b>		
	$\text{Zr}^{97}$	17.0 h	2.2	0.8
	$\text{Nb}^{90}$	16.0 h	1.0	1.0
	$\text{Nb}^{91}$	62 d		0.9, 0.15
	$\text{Nb}^{92}$	10.1 d	1.38, 0.59	1.0
	$\text{Nb}^{92}$	21.6 h	1.2	0.6
	$\text{Nb}^{93}$	<b>100%</b>		
	$\text{Nb}^{94}$	6.6 m	1.3	1.0
	$\text{Nb}^{94}$	>104 y		
	$\text{Nb}^{95}$	87 h		0.2
	$\text{Nb}^{95}$	35 d	0.15	0.77
	$\text{Nb}^{96}$	3 d	1.8	1.0
	$\text{Nb}^{97}$	68 m	1.4	0.8
	$\text{Nb}^{98}$	30 m		
42	$\text{Mo}^{92}$	<b>15.86%</b>		
	$\text{Mo}^{93}$	6.7 h	0.7, 0.3	1.6
	$\text{Mo}^{94}$	<b>9.12%</b>		
	$\text{Mo}^{95}$	<b>15.7%</b>		
	$\text{Mo}^{96}$	<b>16.5%</b>		
	$\text{Mo}^{97}$	<b>9.45%</b>		
	$\text{Mo}^{98}$	<b>23.75%</b>		
	$\text{Mo}^{99}$	67 h	1.4	0.8, 0.4



ATOMIC NUMBER	ATOMIC SPECIES	ABUNDANCE OR HALF-LIFE	PARTICLE ENERGIES (Mev)	QUANTUM ENERGIES
43	<b>Mo<sup>100</sup></b>	<b>9.62%</b>		
	<b>Mo<sup>101</sup></b>	14.6 m	2.2, 1.0	0.9, 0.3
	<i>Tc<sup>92</sup></i>	<i>4.5 m</i>	<i>4.3</i>	<i>1.3</i>
	<b>Tc<sup>94</sup></b>	53 m		0.0334
	<i>Tc<sup>94</sup></i>	<i>25 m</i>	<i>2.47</i>	<i>2.7, 1.8, 1.5, 0.9, 0.4</i>
	<i>Tc<sup>95</sup></i>	<i>56 d</i>	<i>0.4</i>	<i>1.1, 0.8, 0.6, 0.2</i>
	<b>Tc<sup>95</sup></b>	20 h	K	1.0, 0.9, 0.75
	<b>Tc<sup>96</sup></b>	4.3 d	0.6	1.1, 0.8, 0.3
	<b>Tc<sup>97</sup></b>	90 d		0.108
	<b>Tc<sup>97</sup></b>	>100 y		
	<b>Tc<sup>98</sup></b>	2.7 d	1.3	1.0
	<b>Tc<sup>99</sup></b>	6.0 h		0.14
	<b>Tc<sup>99</sup></b>	$9 \times 10^5$ y	0.3	None
	<b>Tc<sup>100</sup></b>	80 s	2.3	0.6
	<b>Tc<sup>101</sup></b>	14 m	1.3	0.3
44	<i>Ru<sup>95</sup></i>	<i>1.65 h</i>	<i>1.1</i>	<i>0.95</i>
	<b>Ru<sup>96</sup></b>	<b>5.68%</b>		
	<i>Ru<sup>97</sup></i>	<i>2.8 d</i>	<i>K</i>	<i>0.23</i>
	<b>Ru<sup>98</sup></b>	<b>2.22%</b>		
	<b>Ru<sup>99</sup></b>	<b>12.81%</b>		
	<b>Ru<sup>100</sup></b>	<b>12.70%</b>		
	<b>Ru<sup>101</sup></b>	<b>16.98%</b>		
	<b>Ru<sup>102</sup></b>	<b>31.34%</b>		
	<b>Ru<sup>103</sup></b>	42 d	0.8, 0.3	0.6
	<b>Ru<sup>104</sup></b>	<b>18.27%</b>		
	<b>Ru<sup>105</sup></b>	4.5 h	1.4	0.7
	<b>Ru<sup>106</sup></b>	1.0 y	0.03	None
45	<i>Rh<sup>100</sup></i>	<i>20 h</i>	<i>3.0, 0.6(—)</i>	<i>1.8, 1.2</i>
	<b>Rh<sup>101</sup></b>	5 d	K	0.35
	<i>Rh<sup>102</sup></i>	<i>210 d</i>	<i>1.13, 1.0(—)</i>	
	<b>Rh<sup>103</sup></b>	<b>100%</b>		
	<b>Rh<sup>103</sup></b>	55 m	0.03	
	<b>Rh<sup>104</sup></b>	4.2 m		0.06
	<b>Rh<sup>104</sup></b>	44 s	2.3	0.9, 0.2
	<b>Rh<sup>105</sup></b>	36 h	0.7	0.3
	<b>Rh<sup>106</sup></b>	30 s	3.5, 2.3	1.25, 0.73, 0.5
46	<i>Pd<sup>100</sup></i>	<i>4.0 d</i>	<i>K</i>	<i>1.8, 0.09</i>
	<i>Pd<sup>101</sup></i>	<i>9 h</i>	<i>2.3</i>	<i>None</i>
	<b>Pd<sup>102</sup></b>	<b>0.8%</b>		
	<i>Pd<sup>103</sup></i>	<i>17 d</i>	<i>K</i>	
	<b>Pd<sup>104</sup></b>	<b>9.3%</b>		
	<b>Pd<sup>105</sup></b>	<b>22.6%</b>		
	<b>Pd<sup>106</sup></b>	<b>27.2%</b>		
	<b>Pd<sup>108</sup></b>	<b>26.8%</b>		
	<b>Pd<sup>109</sup></b>	13 h	1.0	None



ATOMIC NUMBER	ATOMIC SPECIES	ABUNDANCE OR HALF-LIFE	PARTICLE ENERGIES (Mev)	QUANTUM ENERGIES
47	<b>Pd<sup>110</sup></b>	<b>13.5%</b>		
	Pd <sup>111</sup>	26 m	3.5	
	Pd <sup>112</sup>	21 h	0.2	None
	<i>Ag<sup>105</sup></i>	<i>45 d</i>	<i>K</i>	<i>0.6, 0.4, 0.3</i>
	<i>Ag<sup>106</sup></i>	<i>24.5 m</i>	<i>2.0</i>	<i>None</i>
	Ag <sup>106</sup>	8 d	K, 1.2(—)	1.6, 1.0, 0.7
	<b>Ag<sup>107</sup></b>	<b>51.35%</b>		
	Ag <sup>108</sup>	2.3 m	2.8	
	<b>Ag<sup>109</sup></b>	<b>48.65%</b>		
	Ag <sup>110</sup>	24 s	2.6	
	Ag <sup>110</sup>	225 d	1.3, 0.6, 0.4	1.4, 0.9, 0.7
	Ag <sup>111</sup>	7.5 d	1.0	None
	Ag <sup>112</sup>	3.2 h	3.6	0.9
	Ag <sup>113</sup>	5.3 h	2.2	None
48	<b>Cd<sup>106</sup></b>	<b>1.21%</b>		
	Cd <sup>107</sup>	6.7 h	0.32, <i>K</i>	0.8
	<b>Cd<sup>108</sup></b>	<b>0.875%</b>		
	Cd <sup>109</sup>	330 d	<i>K</i>	
	<b>Cd<sup>110</sup></b>	<b>12.39%</b>		
	<b>Cd<sup>111</sup></b>	<b>12.75%</b>		
	<b>Cd<sup>112</sup></b>	<b>24.07%</b>		
	<b>Cd<sup>113</sup></b>	<b>12.26%</b>		
	<b>Cd<sup>114</sup></b>	<b>28.86%</b>		
	Cd <sup>115</sup>	2.33 d	1.25, 0.6	0.6
	Cd <sup>115</sup>	43 d	1.85	0.5
	<b>Cd<sup>116</sup></b>	<b>7.58%</b>		
	Cd <sup>117</sup>	170 m	1.5	
49	<i>In<sup>109</sup></i>	<i>6 h</i>	<i>2</i>	<i>0.5</i>
	<i>In<sup>110</sup></i>	<i>65 m</i>	<i>1.6</i>	
	In <sup>111</sup>	2.7 d		0.25, 0.17
	In <sup>112</sup>	20 m		0.16
	<i>In<sup>112</sup></i>	<i>9 m</i>	<i>1.6</i>	
	In <sup>113</sup>	105 m		0.39
	<b>In<sup>113</sup></b>	<b>4.23%</b>		
	In <sup>114</sup>	48 d		0.19
	In <sup>114</sup>	72 s	2.0	
	In <sup>115</sup>	4.5 h		0.34
	<b>In<sup>115</sup></b>	<b>95.77%</b>		
	In <sup>116</sup>	13 s	2.8	None
	In <sup>116</sup>	54 m	0.85	2.3, 1.3, 1.1, 0.4
	In <sup>117</sup>	117 m	1.73	None
50	<b>Sn<sup>112</sup></b>	<b>0.90%</b>		
	Sn <sup>113</sup>	105 d		
	<b>Sn<sup>114</sup></b>	<b>0.61%</b>		
	<b>Sn<sup>115</sup></b>	<b>0.35%</b>		



ATOMIC NUMBER	ATOMIC SPECIES	ABUNDANCE OR HALF-LIFE	PARTICLE ENERGIES (Mev)	QUANTUM ENERGIES
51	Sn <sup>116</sup>	14.07%		
	Sn <sup>117</sup>	7.54%		
	Sn <sup>118</sup>	23.98%		
	Sn <sup>119</sup>	8.62%		
	Sn <sup>120</sup>	33.03%		
	Sn <sup>121</sup>	27 h	0.4	None
	Sn <sup>122</sup>	4.78%		
	Sn <sup>124</sup>	6.11%		
	Sn <sup>125</sup>	10 m	2.0	0.7
	Sb <sup>118</sup>	3.4 m	3.1	
	Sb <sup>119</sup>	39 h	K	None
	Sb <sup>120</sup>	17 m	1.53	
	Sb <sup>120</sup>	6 d	K, (—)	1.1
	Sb <sup>121</sup>	57.25%		
	Sb <sup>122</sup>	3.5 m		0.14
	Sb <sup>122</sup>	2.8 d	1.9, 1.4	1.0, 0.6
	Sb <sup>123</sup>	42.75%		
	Sb <sup>124</sup>	60 d	2.4, 1.6, 1.0, 0.6	2.0, 1.7, 0.7, 0.6, 0.1
	Sb <sup>124</sup>	21 m		
	Sb <sup>124</sup>	1.3 m	3.2	
52	Sb <sup>125</sup>	2.7 y	0.7, 0.3	0.6
	Sb <sup>127</sup>	92 h	1.1	0.7
	Te <sup>119</sup>	4.5 d	0.5, 0.2	1.4
	Te <sup>120</sup>	0.091%		
	Te <sup>121</sup>	143 d		0.2, 0.16
	Te <sup>121</sup>	5 × 10 <sup>-8</sup> s		0.23
	Te <sup>121</sup>	17 d	K	0.6
	Te <sup>122</sup>	2.49%		
	Te <sup>123</sup>	0.89%		
	Te <sup>124</sup>	4.63%		
	Te <sup>125</sup>	7.01%		
	Te <sup>126</sup>	18.72%		
	Te <sup>127</sup>	9.3 h	0.8	None
	Te <sup>128</sup>	31.72%		
	Te <sup>129</sup>	72 m	1.8	0.8, 0.3
	Te <sup>130</sup>	34.46%		
	Te <sup>131</sup>	25 m		
	Te <sup>132</sup>	77 h	0.36	0.2
	Te <sup>133</sup>	60 m		
	Te <sup>134</sup>	43 m		
53	I <sup>124</sup>	4 d		
	I <sup>125</sup>	56 d	K	None
	I <sup>126</sup>	13.0 d	1.1	0.5
	I <sup>127</sup>	100%		
	I <sup>128</sup>	25 m	2.0, 1.6	0.4
	I <sup>130</sup>	12.6 h	1.0, 0.6	0.7, 0.5, 0.4



ATOMIC NUMBER	ATOMIC SPECIES	ABUNDANCE OR HALF-LIFE	PARTICLE ENERGIES (Mev)	QUANTUM ENERGIES
	$I^{131}$	8.0 d	0.595	0.367, 0.080
	$I^{132}$	2.4 h	2.2, 0.9	1.4, 0.6
	$I^{133}$	22 h	1.4	0.55
	$I^{135}$	6.7 h	1.4, 1.0, 0.5	2.0, 1.27
54	$Xe^{124}$	0.094%		
	$Xe^{126}$	0.088%		
	$Xe^{128}$	1.90%		
	$Xe^{129}$	26.23%		
	$Xe^{130}$	4.07%		
	$Xe^{131}$	21.17%		
	$Xe^{132}$	26.96%		
	$Xe^{133}$	5.3 d	0.34	0.085
	$Xe^{134}$	10.54%		
	$Xe^{135}$	9.2 h	0.93	0.25
	$Xe^{136}$	8.95%		
	$Xe^{137}$	3.8 m	4.0	
	$Xe^{139}$	41 s		
55	$Cs^{132}$	7.1 d	$K (0.6 -)$	0.62
	$Cs^{133}$	100%		
	$Cs^{134}$	2.3 y	0.65	1.35, 0.8, 0.6
	$Cs^{136}$	13.7 d	0.3	1.0
	$Cs^{137}$	37 y	0.55	
	$Cs^{138}$	33 m	2.6	1.2
56	$Ba^{130}$	0.101%		
	$Ba^{131}$	12 d	$K$	1.7, 0.5, 0.22
	$Ba^{132}$	0.097%		
	$Ba^{133}$	38 h		0.30
	$Ba^{133}$	30 y	$K$	0.36
	$Ba^{134}$	2.42%		
	$Ba^{135}$	28.7 h		0.34
	$Ba^{135}$	6.59%		
	$Ba^{136}$	7.81%		
	$Ba^{137}$	2.6 m	0.6(—)	0.66
	$Ba^{137}$	11.32%		
	$Ba^{138}$	71.66%		
	$Ba^{139}$	84 m	2.3	1.0, 0.16
	$Ba^{140}$	308 h	1.0, 0.4	0.5
57	$La^{135}$	19 h	$K$	0.9
	$La^{136}$	2.1 h	0.8	None
	$La^{138}$	0.089%		
	$La^{139}$	99.911%		
	$La^{140}$	40 h	2.1, 1.4, 0.9	2.3, 1.6, 0.9, 0.5
	$La^{141}$	3.7 h	2.9	
58	$Ce^{136}$	0.193%		
	$Ce^{137}$	36 h	$K$	0.75, 0.3



ATOMIC NUMBER	ATOMIC SPECIES	ABUNDANCE OR HALF-LIFE	PARTICLE ENERGIES (Mev)	QUANTUM ENERGIES
	<b>Ce<sup>138</sup></b>	<b>0.250%</b>		
	Ce <sup>139</sup>	140 d	K	1.8, 0.2
	<b>Ce<sup>140</sup></b>	<b>88.48%</b>		
	Ce <sup>141</sup>	29 d	0.6	0.2
	<b>Ce<sup>142</sup></b>	<b>11.07%</b>		
	Ce <sup>143</sup>	34 h	1.3	0.5
	Ce <sup>144</sup>	280 d	0.3	None
59	Pr <sup>140</sup>	3.5 m	2.5	
	<b>Pr<sup>141</sup></b>	<b>100%</b>		
	Pr <sup>142</sup>	19.3 h	2.1	1.7
	Pr <sup>143</sup>	13.8 d	1.0	None
	Pr <sup>144</sup>	17.5 m	3.05	1.2, 0.2
60	Nd <sup>141</sup>	2.4 h	0.8, K	1.0
	<b>Nd<sup>142</sup></b>	<b>27.13%</b>		
	<b>Nd<sup>143</sup></b>	<b>12.20%</b>		
	<b>Nd<sup>144</sup></b>	<b>23.87%</b>		
	<b>Nd<sup>145</sup></b>	<b>8.30%</b>		
	<b>Nd<sup>146</sup></b>	<b>17.18%</b>		
	Nd <sup>147</sup>	11 d	0.9, 0.4	0.6
	<b>Nd<sup>148</sup></b>	<b>5.72%</b>		
	Nd <sup>149</sup>	1.8 h	1.6	
	<b>Nd<sup>150</sup></b>	<b>5.60%</b>		
	<b>Nd<sup>150?</sup></b>	$5 \times 10^{10}$ y	0.011	Naturally radioactive
61	Pm <sup>143</sup>	300 d	K	0.7
	Pm <sup>147</sup>	3.8 y	0.22	None
	Pm <sup>148</sup>	5.3 d	2.3	0.8
	Pm <sup>149</sup>	47 h	1.1	0.25
62	<b>Sm<sup>144</sup></b>	<b>3.16%</b>		
	<b>Sm<sup>147</sup></b>	<b>15.07%</b>		
	<b>Sm<sup>148</sup></b>	<b>11.27%</b>		
	<b>Sm<sup>149</sup></b>	<b>13.84%</b>		
	<b>Sm<sup>150</sup></b>	<b>7.47%</b>		
	Sm <sup>151</sup>	20 y	0.06	
	<b>Sm<sup>152</sup></b>	<b>26.63%</b>		
	Sm <sup>152</sup>	$1.0 \times 10^{12}$ y, $\alpha$	2.1 $\alpha$	Naturally radioactive
	Sm <sup>153</sup>	47 h	0.78	0.6, 0.1
	<b>Sm<sup>154</sup></b>	<b>22.53%</b>		
	Sm <sup>155</sup>	25 m	1.9	0.3
63	Eu <sup>147</sup>	48 d		
	Eu <sup>149</sup>	14 d		
	Eu <sup>150</sup>	27 h		
	<b>Eu<sup>151</sup></b>	<b>47.77%</b>		
	Eu <sup>152</sup>	9.2 h	1.88	0.72, 0.16, 0.12
	Eu <sup>152</sup>	Long	0.75	
	<b>Eu<sup>153</sup></b>	<b>52.23%</b>		



ATOMIC NUMBER	ATOMIC SPECIES	ABUNDANCE OR HALF-LIFE	PARTICLE ENERGIES (Mev)	QUANTUM ENERGIES
64	Eu <sup>154</sup>	7 y	0.9	1.2, 0.4, 0.3, 0.12
	Eu <sup>155</sup>	2.5 y	0.2	0.08
	Gd <sup>152</sup>	0.20%	K	0.27
	Gd <sup>153</sup>	155 d		
	Gd <sup>154</sup>	2.15%		
	Gd <sup>155</sup>	14.78%		
	Gd <sup>156</sup>	20.59%		
	Gd <sup>157</sup>	15.71%		
	Gd <sup>158</sup>	24.78%		
	Gd <sup>160</sup>	21.79%		
	Gd <sup>161</sup>	18 h	0.85	0.3
	Gd <sup>161</sup>	4.5 m	1.5	0.37
65	Tb <sup>154</sup>	17.2 h	2.6, 0.2	1.4
	Tb <sup>155</sup>	1 y	K	
	Tb <sup>159</sup>	100%		
	Tb <sup>160</sup>	3.9 h		
	Tb <sup>160</sup>	73 d	0.9, 0.55	1.15, 0.3, 0.2, 0.1
	Tb <sup>161</sup>	5.5 d	0.5	1.3
66	Dy <sup>156</sup>	0.0524%	1.25, 0.9, 0.4	0.7, 0.4, 0.1
	Dy <sup>158</sup>	0.0902%		
	Dy <sup>160</sup>	2.294%		
	Dy <sup>161</sup>	18.88%		
	Dy <sup>162</sup>	25.53%		
	Dy <sup>163</sup>	24.97%		
	Dy <sup>164</sup>	28.18%		
	Dy <sup>165</sup>	145 m		
67	Ho <sup>163</sup>	7 d	K	
	Ho <sup>164</sup>	35 m	0.7	
	Ho <sup>165</sup>	100%		
	Ho <sup>166</sup>	27 h	1.8	
68	Er <sup>162</sup>	0.1%	0.3	None
	Er <sup>164</sup>	1.5%		
	Er <sup>165</sup>	1.1 m		
	Er <sup>166</sup>	32.9%		
	Er <sup>167</sup>	24.4%		
	Er <sup>168</sup>	26.9%		
	Er <sup>169</sup>	9.4 d		
	Er <sup>170</sup>	14.2%		
	Er <sup>171</sup>	7.5 h		
			1.5, 1.0, 0.7	0.8, 0.3, 0.1
69	Tm <sup>166</sup>	7.7 h	2.1, 0.2	1.5
	Tm <sup>167</sup>	9 d	K	0.9, 0.2
	Tm <sup>169</sup>	10 <sup>-6</sup> s, $\gamma$		
	Tm <sup>169</sup>	100%		
	Tm <sup>170</sup>	127 d	1.0	0.8



ATOMIC NUMBER	ATOMIC SPECIES	ABUNDANCE OR HALF-LIFE	PARTICLE ENERGIES (Mev)	QUANTUM ENERGIES
70	$\text{Tm}^{171}$	$2.5 \times 10^{-6} \text{ s}, \alpha$		0.1
	$\text{Tm}^{171}$	500 d	0.1	
	$\text{Yb}^{168}$	0.06%		
	$\text{Yb}^{169}$	33 d	K	0.4, 0.2
	$\text{Yb}^{170}$	4.21%		
	$\text{Yb}^{171}$	14.26%		
	$\text{Yb}^{172}$	21.49%		
	$\text{Yb}^{173}$	17.02%		
	$\text{Yb}^{174}$	29.58%		
	$\text{Yb}^{175}$	99 h	0.5, 0.13	0.35
	$\text{Yb}^{176}$	13.38%		
71	$\text{Yb}^{177}$	2.4 h	1.3	
	$\text{Lu}^{170}$	2.15 d	1.7	1.5
	$\text{Lu}^{171}$	9 d	0.17	0.8
	$\text{Lu}^{175}$	97.5%		
	$\text{Lu}^{176}$	2.5%, $7.3 \times 10^{10} \text{ y}$	0.3	0.3, naturally radio- active
	$\text{Lu}^{176}$	3.7 h	1.1	None
72	$\text{Lu}^{177}$	6.8 h	0.45	1.3, 0.2
	$\text{Hf}^{174}$	0.18%		
	$\text{Hf}^{175}$	70 d	K	1.5, 0.3
	$\text{Hf}^{176}$	5.30%		
	$\text{Hf}^{177}$	18.47%		
	$\text{Hf}^{178}$	27.10%		
	$\text{Hf}^{179}$	13.84%		
	$\text{Hf}^{180}$	35.11%		
	$\text{Hf}^{181}$	50 d	0.46	0.5, 0.35, 0.13
73	$\text{Ta}^{176}$	8 h	K	1.7
	$\text{Ta}^{177}$	2.7 d	K	
	$\text{Ta}^{180}$	8 h	K	
	$\text{Ta}^{181}$	$2.0 \times 10^{-6} \text{ s}, \gamma$		0.472, 0.13
	$\text{Ta}^{181}$	100%		
	$\text{Ta}^{182}$	117 d	1.0	1.2, 1.1, 0.2, 0.15
74	$\text{W}^{180}$	0.122%		
	$\text{W}^{181}$	140 d	K	1.8, 0.14
	$\text{W}^{182}$	25.77%		
	$\text{W}^{183}$	14.24%		
	$\text{W}^{184}$	30.68%		
	$\text{W}^{185}$	73 d	0.42	None
	$\text{W}^{186}$	29.17%		
	$\text{W}^{187}$	24.1 h	1.3, 0.6	0.7, 0.5, 0.13
75	$\text{Re}^{182}$	64 h	K	1.52, 0.23
	$\text{Re}^{184}$	50 d	0.2, K	1.0, 0.17
	$\text{Re}^{185}$	37.07%		



ATOMIC NUMBER	ATOMIC SPECIES	ABUNDANCE OR HALF-LIFE	PARTICLE ENERGIES (Mev)	QUANTUM ENERGIES
76	Re <sup>186</sup>	93 h	1.07	
	Re <sup>187</sup>	$0.6 \times 10^{-6}$ s, $\gamma$		
	Re <sup>187</sup>	<b>62.93%</b> , $4 \times 10^{12}$ y	0.043	Naturally radioactive
	Re <sup>188</sup>	19 h	2.0	1.4, 0.9, 0.6, 0.5, 0.18
	Os <sup>184</sup>	<b>0.018%</b>		
	Os <sup>185</sup>	97 d	K	0.75
	Os <sup>186</sup>	<b>1.59%</b>		
	Os <sup>187</sup>	<b>1.64%</b>		
	Os <sup>188</sup>	<b>13.3%</b>		
	Os <sup>189</sup>	<b>16.1%</b>		
	Os <sup>190</sup>	<b>26.4%</b>		
	Os <sup>191</sup>	15 d	0.15	0.12, 0.039
	Os <sup>192</sup>	<b>41.0%</b>		
	Os <sup>193</sup>	32 h	1.5	1.6
77	Ir <sup>190</sup>	10.7 d	K	0.25
	Ir <sup>191</sup>	<b>38.5%</b>		
	Ir <sup>192</sup>	1.5 m, $\gamma$		0.06
	Ir <sup>192</sup>	70 d	0.7	0.6, 0.46, 0.30
	Ir <sup>193</sup>	<b>61.5%</b>		
	Ir <sup>194</sup>	19 h	2.2	1.35, 0.4
78	Pt <sup>191</sup>	3 d	K	1.5, 0.6
	Pt <sup>192</sup>	<b>0.78%</b>		
	Pt <sup>193</sup>	4.3 d	K	1.5, 0.18
	Pt <sup>194</sup>	<b>32.8%</b>		
	Pt <sup>195</sup>	<b>33.7%</b>		
	Pt <sup>196</sup>	<b>24.4%</b>		
	Pt <sup>196</sup>	80 m, $\gamma$		0.34
	Pt <sup>197</sup>	18 h	0.7	
	Pt <sup>197</sup>	3.3 d, $\gamma$		
	Pt <sup>198</sup>	<b>7.3%</b>		
	Pt <sup>199</sup>	31 m	1.8	
79	Au <sup>192</sup>	4.7 h	K	2.3
	Au <sup>193</sup>	15.8 h	K	
	Au <sup>194</sup>	39.5 h	K	1.5, 0.3
	Au <sup>195</sup>	185 d	K	1.6, 0.19
	Au <sup>196</sup>	14 h		
	Au <sup>196</sup>	5.6 d	0.4	0.3, 0.14
	Au <sup>197</sup>	<b>100%</b>		
	Au <sup>197</sup>	7.5 s, $\gamma$		0.27
	Au <sup>198</sup>	2.7 d	0.96, 0.60	0.41, 0.16
	Au <sup>199</sup>	3.3 d	0.38	0.3
80	Hg <sup>196</sup>	<b>0.15%</b>		
	Hg <sup>197</sup>	23 h	K	0.16, 0.13
	Hg <sup>197</sup>	64 h	K	0.07



ATOMIC NUMBER	ATOMIC SPECIES	ABUNDANCE OR HALF-LIFE	PARTICLE ENERGIES (Mev)	QUANTUM ENERGIES
81	Hg <sup>198</sup>	0.3 × 10 <sup>-6</sup> s, γ		0.4
	Hg <sup>198</sup>	10.1%		
	Hg <sup>199</sup>	17.0%		
	Hg <sup>200</sup>	23.3%		
	Hg <sup>202</sup>	29.6%		
	Hg <sup>203</sup>	46 d	0.2	0.3
	Hg <sup>204</sup>	6.7%		
	Hg <sup>205</sup>	5.5 m	1.62	
	Tl <sup>198</sup>	1.8 h	K	1.3
	Tl <sup>199</sup>	7 h	K	1.5
	Tl <sup>200</sup>	27 h	K	0.4
	Tl <sup>202</sup>	11.8 d	K	0.4
	Tl <sup>203</sup>	29.1%		
	Tl <sup>204</sup>	2.7 y	0.8	None
	Tl <sup>205</sup>	70.9%		
82	Tl <sup>206</sup>	4.2 m	1.7	None
	Tl <sup>207</sup> (AcC <sup>11</sup> )	4.76 m	1.47	
	Tl <sup>208</sup> (ThC <sup>11</sup> )	3.1 m	1.72	2.62
	Tl <sup>209</sup>	2.2 m	1.8	
	Tl <sup>210</sup> (RaC <sup>11</sup> )	1.32 m	1.80	
	Pb <sup>203</sup>	52 h	K	0.45
	Pb <sup>204</sup>	1.5%		
	Pb <sup>204</sup>	68 m, γ		1.1
	Pb <sup>206</sup>	23.6%		
	Pb <sup>207</sup>	22.6%		
	Pb <sup>208</sup>	52.3%		
	Pb <sup>209</sup>	3.3 h	0.7	None
	Pb <sup>210</sup> (RaD)	22 y	0.025	0.047
	Pb <sup>211</sup> (AcB)	36 m	1.4, 0.5	0.8
	Pb <sup>212</sup> (ThB)	10.6 h	0.36	
	Pb <sup>214</sup> (RaB)	26.8 m	0.65	
83	Bi <sup>204</sup>	12 h	K	
	Bi <sup>206</sup>	6.4 d	K	0.74
	Bi <sup>209</sup>	100%		
	Bi <sup>210</sup> (RaE)	5 d	1.17	None
	Bi <sup>211</sup> (AcC)	2.16 m, α	6.62α	
	Bi <sup>212</sup> (ThC)	60.5 m, α	6.08α	
	Bi <sup>213</sup>	47 m	1.3	
	Bi <sup>214</sup> (RaC)	19.7 m, α	5.5α	1.8
84	Po <sup>206</sup>	9 d, also α	K 5.2α	0.8
	Po <sup>207</sup>	5.7 h	K	1.3
	Po <sup>208</sup>	3 y, α	5.14α	None
	Po <sup>210</sup>	138 d, α	5.3α	0.7 weak
	Po <sup>211</sup> (AcC')	5 × 10 <sup>-3</sup> s, α	7.434α	
	Po <sup>212</sup> (ThC')	3.0 × 10 <sup>-7</sup> s, α	8.776α	



ATOMIC NUMBER	ATOMIC SPECIES	ABUNDANCE OR HALF-LIFE	PARTICLE ENERGIES (Mev)	QUANTUM ENERGIES
	Po <sup>213</sup>	$4.2 \times 10^{-6}$ s, $\alpha$	8.336 $\alpha$	
	Po <sup>214</sup> (RaC')	$1.5 \times 10^{-4}$ s, $\alpha$	7.68 $\alpha$	
	Po <sup>215</sup> (AcA)	$1.8 \times 10^{-3}$ s, $\alpha$	7.36 $\alpha$	
	Po <sup>216</sup> (ThA)	0.16 s, $\alpha$	6.77 $\alpha$	
	Po <sup>218</sup> (RaA)	3.05 m, $\alpha$	6.0 $\alpha$	
85	At <sup>210</sup>	8.3 h	K	1.0
	At <sup>211</sup>	7.5 h, 40% $\alpha$	K, 5.89 $\alpha$	
	At <sup>215</sup>	$10^{-4}$ s, $\alpha$	8.00 $\alpha$	
	At <sup>216</sup>	$3 \times 10^{-4}$ s, $\alpha$	7.79 $\alpha$	
	At <sup>217</sup>	0.018 s, $\alpha$	7.02 $\alpha$	
86	Au <sup>219</sup>	3.92 s, $\alpha$	6.82 $\alpha$	
	Tu <sup>219</sup>	54.5 s, $\alpha$	6.28 $\alpha$	
	Rn <sup>222</sup>	3.825 d, $\alpha$	5.49 $\alpha$	
87	Fr <sup>223</sup> (AcK)	21 m	1.2	0.09
88	Ra <sup>223</sup> (AcX)	11.2 d, $\alpha$	5.7	
	Ra <sup>224</sup> (ThX)	3.64 d, $\alpha$	5.68 $\alpha$	
	Ra <sup>225</sup>	14.8 d	0.1	
	Ra <sup>226</sup>	1622 y, $\alpha$	4.79 $\alpha$	0.19, naturally radio- active
	Ra <sup>228</sup> (MsTh)	6.7 y	0.05	Naturally radioactive
89	Ac <sup>225</sup>	10 d, $\alpha$	5.8 $\alpha$	
	Ac <sup>227</sup>	21 y	0.01, also 4.9 $\alpha$	0.04
	Ac <sup>228</sup> (McTh <sub>2</sub> )	6.13 h	1.55	Naturally radioactive
90	Th <sup>227</sup> (RdAc)	18.6 d, $\alpha$	6.05 $\alpha$ and others	Naturally radioactive
	Th <sup>228</sup> (RdTh)	1.9 y	5.42	Naturally radioactive
	Th <sup>229</sup>	7000 y, $\alpha$	5.0 $\alpha$ and others	
	Th <sup>230</sup> (Io)	$8.0 \times 10^4$ y, $\alpha$	4.66 $\alpha$	Naturally radioactive
	Th <sup>231</sup> (UY)	25.5 h	0.2	0.035, naturally radioactive
	Th <sup>232</sup>	100%		
	Th <sup>233</sup>	23.5 m	1.2	None
	Th <sup>234</sup> (UX <sub>1</sub> )	24.1 d	0.2	0.09, naturally radio- active
91	Pa <sup>230</sup>	17.7 d	1.1	0.9
	Pa <sup>231</sup>	$3.4 \times 10^4$ y, $\alpha$	5.01 $\alpha$ , and others	0.3, 0.095, naturally radioactive
	Pa <sup>232</sup>	1.32 d	0.3	1.0
	Pa <sup>233</sup>	27 d	0.4	0.3, 0.084
	Pa <sup>234</sup> (UZ)	6.7 h	1.55, 0.56	0.70
	Pa <sup>234</sup> (UX <sub>2</sub> )	1.14 m	2.3, 1.52	0.8
92	U <sup>233</sup>	$1.6 \times 10^5$ y, $\alpha$	4.82 $\alpha$	0.3, 0.08, 0.04
	U <sup>234</sup>	0.0051%		



ATOMIC NUMBER	ATOMIC SPECIES	ABUNDANCE OR HALF-LIFE	PARTICLE ENERGIES (Mev)	QUANTUM ENERGIES
	(U <sub>II</sub> ) <b>U<sup>235</sup></b>	$2.35 \times 10^5$ y, $\alpha$ <b>0.71%</b>	$4.76\alpha$	Naturally radioactive
	(AcU) <b>U<sup>237</sup></b>	$8.9 \times 10^8$ y, $\alpha$ 6.8 d	$4.56\alpha$ 0.25	Naturally radioactive 0.26, 0.24, 0.06
	<b>U<sup>238</sup></b>	<b>99.28%</b>		
	(U <sub>I</sub> ) <b>U<sup>239</sup></b>	$4.5 \times 10^8$ y, $\alpha$ 23.0 m	$4.18\alpha$ 2.06, 1.12	Naturally radioactive 0.92, 0.08
93	<b>Np<sup>235</sup></b>	435 d	K	
	<b>Np<sup>236</sup></b>	22 h	0.5	
	<b>Np<sup>237</sup></b>	$2.2 \times 10^6$ y, $\alpha$	4.77	
	<b>Np<sup>238</sup></b>	2.1 d	1.39, 0.22	1.2, 0.075
	<b>Np<sup>239</sup></b>	2.33 d	0.68, 0.33, 0.09	0.27, 0.23, 0.06
94	<b>Pu<sup>238</sup></b>	92 y, $\alpha$	$5.51\alpha$	
	<b>Pu<sup>239</sup></b>	$2.411 \times 10^4$ y, $\alpha$	$5.15\alpha$	
95	<b>Am<sup>239</sup></b>	12 h	K	0.29
	<b>Am<sup>240</sup></b>	50 h	K	1.3
	<b>Am<sup>241</sup></b>	490 y, $\alpha$	$5.48\alpha$	0.06
	<b>Am<sup>242</sup></b>	16 h	0.8	
	<b>Am<sup>242</sup></b>	400 y, $\alpha$		
96	<b>Cm<sup>240</sup></b>	26.8 d, $\alpha$	$6.26\alpha$	
	<b>Cm<sup>242</sup></b>	150 d, $\alpha$	$6.08\alpha$	



## APPENDIX 3

### COMMONLY USED RADIOELEMENTS

The elements available from the Isotopes Division of the Atomic Energy Commission can all be classed as in common use. We list here those which are used to a considerable degree. The letters *cyc* follow any isotope which is available only if charged particle bombardment can be used.

ELEMENT	HALF-LIFE	ELECTRON ENERGIES	QUANTUM ENERGIES
H <sup>3</sup>	12.4 y *	0.019	None
C <sup>14</sup>	5000 y	0.145	None
Na <sup>22</sup> <i>cyc</i>	3.0 y	0.6	1.3
Na <sup>24</sup>	14.8 h *	1.4	2.8, 1.4
P <sup>32</sup>	14.3 d *	1.7	None
S <sup>35</sup>	88 d	0.17	None
Cl <sup>38</sup> <i>cyc</i>	37 m *	4.9, 2.9, 1.2	2.2, 1.6
K <sup>42</sup>	12.4 h	3.6, 2.0	1.6
Ca <sup>45</sup>	180 d	0.25	None
Mn <sup>56</sup> <i>cyc</i>	2.6 h	2.8, 1.0, 0.7	2.1, 1.8, 0.8
Fe <sup>55</sup>	4 y	K	
Fe <sup>59</sup>	47 d	0.46, 0.26	1.3, 1.1
Co <sup>56</sup> <i>cyc</i>	72 d	1.5	3.2, 2.5, 2.0, 1.7, 1.3, 0.8
Co <sup>60</sup>	5.3 y	0.3	1.16, 1.32
Cu <sup>64</sup>	12.8 h	0.66, 0.58	None
As <sup>76</sup>	26.8 h	3.0, 2.4, 1.2	1.7, 1.2, 0.6
Br <sup>82</sup>	34 h	0.7	0.65
I <sup>131</sup>	8.0 d	0.7	0.4
Au <sup>198</sup>	2.7 d	0.96	0.4

\* y = year(s); h = hour(s); d = day(s); m = minute(s).



## APPENDIX 4

### ABSORPTION OF BETA RAYS

To understand the absorption of the continuous beta rays emitted by a radioactive substance two relations need to be known. The first is the energy distribution of the electrons themselves, and the second is the relation between the energy of an electron and the thickness of absorber which it will traverse. Neither of these is particularly simple if one aims at very rigorous consideration of absorption, but both can be approximated so as to be very useful in designing experimental equipment and technique.

We have stated in the text that beta rays have a maximum energy for any one element. Suppose that this is  $E_m$  Mev. Also suppose that the probability of emission of electrons of energies between the values  $E$  and  $E + dE$  is  $W$ . Then the Fermi theory gives the following expression for  $W$ .

$$W = A(E)^{1/2}(1 + 2E)(1 + E)^{1/2}(E_m - E)^2 dE \quad (1)$$

This is not exact but is within 2 per cent. In this formula  $A$  is a constant for any one element. To the degree of approximation usually needed for radioactive tracer work this can be replaced by

$$W = B(E)(3 + 2E)(E_m - E)^2 dE \quad (2)$$

This can be relied on to about 15 per cent for values of  $E$  less than 2 Mev. The greatest deviation from this formula is introduced by the method of supporting the radioactive source. If the support is thick there is a considerable amount of scattering of the electrons by the support, and this changes the energy distribution. In the above equation  $B$  is again a constant for any one element.

In Chapter 7 we briefly discussed the absorption of beta rays and gave a rough beta-ray formula. This is approximately true for all kinds of absorbing material, but if aluminum is used a rather better accuracy can be obtained by using an empirical relation first given by Feather and improved by Widdowson and Champion. This is

$$R = 0.536E - 0.165 \quad (3)$$

where  $R$  is the range, or thickness just penetrable, measured in grams per square centimeter. If the reader is prepared to use these two formulas he can get a reasonable idea of the relative counts for different thicknesses of absorbing material.

Where two groups of beta rays are found the correct procedure is to treat each group as having its own separate maximum energy and add the results. Actually a practical average can be taken by treating the element as if it had one maximum energy which is the average of the two most energetic groups.



## APPENDIX 5

### ABSORPTION OF GAMMA RAYS

If a series of absorbers is placed in the path of a parallel beam of gamma rays the absorption takes place exponentially according to the relation

$$\frac{I}{I_0} = e^{-\tau x} \quad \text{or} \quad \ln I_0 - \ln I = \tau x$$

where  $I$  is the ionization current or number of secondary electrons counted at a thickness of absorber  $x$ , and  $I_0$  is the corresponding quantity with no absorber other than enough to give equilibrium ionization.  $\tau$  is called the absorption coefficient.

In the following tables we give the absorption coefficients for several different substances and a large range of energy values. In addition we present a table of half-value thicknesses in aluminum and copper which may help in a quick rough determination of gamma-ray energies. Approximately, brass is the same as copper.

GAMMA-RAY		ABSORPTION COEFFICIENT				
ENERGY						
(Mev)	<i>Carbon</i>	<i>Water</i>	<i>Aluminum</i>	<i>Copper</i>	<i>Lead</i>	
0.25	0.26	0.124	0.29	0.91		
0.50	0.20	0.095	0.22	0.70	1.7	
0.75	0.17		0.19	0.58		
1.00	0.15	0.069	0.16	0.50	0.80	
1.25	0.13		0.146	0.45		
1.50	0.12		0.132	0.41		
1.75	0.114		0.122	0.38		
2.00	0.106		0.115	0.35		
2.50	0.087	0.043	0.105	0.33	0.475	
3.00	0.083		0.100	0.32		
3.50	0.078		0.095	0.31		
4.00	0.069		0.086	0.30		
4.50			0.078	0.28		
5.00		0.030	0.075	0.27	0.480	
5.50			0.073	0.28		
6.00			0.071	0.28		
7.00			0.068	0.30		
8.00			0.065	0.30		
9.00			0.063	0.31		
10.00		0.022	0.061	0.31	0.61	



GAMMA-RAY  
ENERGY  
(Mev)

ABSORPTION COEFFICIENT

<i>Carbon</i>	<i>Water</i>	<i>Aluminum</i>	<i>Copper</i>	<i>Lead</i>
		0.061	0.32	
	0.017	0.054	0.32	
		0.058	0.34	
	0.015	0.061	0.38	1.02

HALF-VALUE THICKNESS FOR DIFFERENT ENERGIES

<i>Aluminum</i>		<i>Copper</i>	
$x_{1/2}$ (cm)	<i>E</i> (Mev)	$x_{1/2}$ (cm)	<i>E</i> (Mev)
2.0	0.15	0.50	0.12
3.0	0.47	0.75	0.25
4.0	0.92	1.00	0.52
5.0	1.40	1.50	1.20
6.0	2.00	2.00	2.10
7.0	2.95	2.10	2.50
8.0	4.00	2.30	3.80



## APPENDIX 6

### MASSES

Below is a very incomplete table of masses which, in a few years time will be practically comprehensive. These masses are accurate up to about  $\text{Si}^{28}$ , but for heavier elements the accuracy is considerably less. The values up to  $\text{F}^{19}$  are taken from an article by Tollestrup, Fowler and Lauritsen (*Phys. Rev.*, **78**, 374, 1950), supplemented by data on boron taken by Bateson. The neon isotopes are taken from Zucker and Watson (*Phys. Rev.*, **78**, 14, 1950); the remainder, up to  $\text{Ca}^{42}$ , are based on transmutation data taken by Motz, Sailor, Wyly, Davison, and Worth. The heaviest values are founded on the work of Duckworth, Johnson, Preston, and Woodcock (*Phys. Rev.*, **78**, 386, 1950), plus transmutation data by Martin and Bateson.

Element	Mass	Element	Mass
$n$	1.00898	$\text{Si}^{28}$	27.9855
$\text{H}^1$	1.00814	$\text{Si}^{29}$	28.9855
$\text{H}^2$	2.01472	$\text{Si}^{30}$	29.9831
$\text{H}^3$	3.01693	$\text{P}^{31}$	30.9820
$\text{He}^3$	3.01695	$\text{S}^{32}$	31.9807
$\text{He}^4$	4.00391	$\text{S}^{33}$	32.9813
$\text{Li}^6$	6.01704	$\text{S}^{34}$	33.9775
$\text{Li}^7$	7.01824	$\text{Cl}^{35}$	34.9787
$\text{Be}^9$	9.01510	$\text{S}^{36}$	34.9778
$\text{B}^{10}$	10.01617	$\text{A}^{36}$	35.9781
$\text{B}^{11}$	11.01293	$\text{Cl}^{37}$	36.9775
$\text{C}^{12}$	12.00390	$\text{A}^{38}$	37.9744
$\text{C}^{13}$	13.00755	$\text{K}^{39}$	38.9762
$\text{N}^{14}$	14.00756	$\text{A}^{40}$	39.9756
$\text{N}^{15}$	15.0049	$\text{Ca}^{40}$	39.9754
$\text{O}^{16}$	16.0000	$\text{K}^{41}$	40.9740
$\text{O}^{17}$	17.0045	$\text{Ca}^{42}$	41.9712
$\text{O}^{18}$	18.0049	$\text{Sc}^{45}$	44.9669
$\text{F}^{19}$	19.0045	$\text{Cr}^{50}$	49.9602
$\text{Ne}^{20}$	19.9989	$\text{Cr}^{52}$	51.9571
$\text{Ne}^{21}$	21.0006	$\text{Fe}^{54}$	53.9566
$\text{Ne}^{22}$	21.9985	$\text{Mn}^{55}$	54.9547
$\text{Na}^{23}$	22.9970	$\text{Fe}^{56}$	55.9523
$\text{Mg}^{24}$	23.9924	$\text{Ni}^{58}$	57.9531
$\text{Mg}^{25}$	25.9936	$\text{Co}^{59}$	58.9513
$\text{Mg}^{26}$	26.9919	$\text{Ni}^{60}$	59.9488
$\text{Al}^{27}$	26.9898		



## APPENDIX 7

### ENERGY AND RANGE RELATIONSHIPS FOR FAST CHARGED PARTICLES

We give here two tables relating the energy and range of various charged particles. These tables are intended for the following purposes: general information, quick calculation of the necessary target thickness in various cases, rough estimation of the range of a beam of particles, and estimation of the range of some group of particles evolved in a reaction. For serious measurement of nuclear energy levels the method of procedure given in the invaluable review article by Livingston and Bethe (*Reviews of Modern Physics*, April 1937) should be followed.

A short word about nuclear energetics can be put in here. If a bombarding particle of atomic mass  $M_B$  has the energy  $E_B$  and produces a transmutation in which a nucleus of mass  $M_N$  and a particle of mass  $M_P$ , energy  $E_P$ , at an angle  $\theta$  to the original direction of bombardment are formed, then if  $Q$  is the appropriate nuclear energy change there is a relation

$$M_N Q = (M_N + M_P)E_P - (M_N - M_B)E_B - 2(M_B M_P E_B E_P)^{1/2} \cos \theta$$

which holds and which can be used to determine any one of the unknown quantities. In the above it is assumed that only one product nucleus and one particle are formed. It is also assumed that the bombarding particle is captured. It is of interest that if the *maximum* value of  $E_B$  is considered then the formula also holds even if the bombarding particle is not captured, for where  $E_B$  is greatest the bombarding particle and the product nucleus move off together. All the above results from the application of the laws of conservation of momentum and energy to the process. Energies are in Mev.

In the tables we give *mean ranges*. The reason for specifying a particular type of range is that there is an element of chance in the penetration of a charged particle through matter. It may happen that a particle follows a path which involves a less than ordinary number of collisions that cause energy loss. The particle will therefore not lose all its kinetic energy until it has traveled farther. Its range is thus abnormally great. It is easy to see that this element of chance will cause a spread in the measured ranges of particles which have initially the same energy. This spread is called *straggling*. As straggling follows a definite law, it is quite possible to measure a quantity which can be related quite accurately to the initial energy. There are several such quantities, of which we select two, the "extrapolated numbers range" and the "mean range." The mean range is the range reached by one-half the particles; the extrapolated numbers range is obtained by plotting a graph of absorption versus numbers and extrapolating the steepest tangent to cut the axis of zero number. The two differ by about 2 per cent for protons and deuterons and 1 per cent for alpha particles. In all cases, of course, the mean range is less. For a thorough consideration of these two ranges the reader is again referred to the article by Livingston and Bethe.



## EQUIVALENT THICKNESSES OF ABSORBERS

In actual work, air is rarely used as the absorber. We here give the thickness in milligrams per square centimeter of surface area of a few standard absorbers which is equivalent to 1 cm. of air.

Al 1.53      Cu 2.10      Ag 2.72      Au 3.77

## ENERGY-RANGE RELATIONS

All ranges are in centimeters air equivalent, energies in Mev

ENERGY	RANGE				
	<i>Proton</i>	<i>Deuteron</i>	<i>H<sup>3</sup></i>	<i>He<sup>3</sup></i>	<i>α Particle</i>
0.2	0.29	0.28	0.24	0.16	0.17
0.4	0.65	0.59	0.57	0.26	0.27
0.6	1.10	0.90	0.87	0.35	0.38
0.8	1.65	1.30	1.20	0.45	0.47
1.0	2.30	1.72	1.53	0.55	0.57
1.2	3.06	2.19	1.95	0.63	0.66
1.4	3.91	2.70	2.37	0.74	0.74
1.6	4.88	3.30	2.83	0.85	0.84
1.8	5.92	3.92	3.30	0.96	0.94
2.0	7.20	4.61	3.90	1.09	1.05
2.5	10.40	6.51	5.22	1.46	1.35
3.0	14.10	8.78	6.90	1.86	1.70
3.5	18.30	11.32	8.79	2.35	2.08
4.0	23.10	14.40	10.81	2.87	2.49
4.5	28.30	17.30	13.17	3.38	2.97
5.0	33.90	20.80	15.60	4.10	3.48
6.0	46.7	28.2	21.6	5.50	4.52
7.0	61.2	36.6	27.5	7.09	5.90
8.0	77.3	46.2	34.8	8.80	7.35
9.0	95.3	56.6	42.3	10.60	8.89
10.0	114.8	67.8	50.2	12.85	10.55
11.0	136.1	80.2	59.5	15.2	12.40
12.0	150.4	93.4	69.3	18.0	14.18
13.0	183.1	107.4	79.0	19.8	16.24
14.0	209.1	122.4	90.3	22.3	18.35
15.0	238.5	138.2	101.7	24.3	21.17
20.0		229.6	169	39.5	32.5
25.0		343	260	62.5	51.0
30.0		477	344	87	71.0
35.0				114	92.5
40.0				144	115.7



## RANGE-ENERGY RELATIONS

RANGE	ENERGY				
	<i>Proton</i>	<i>Deuteron</i>	$H^3$	$He^3$	$\alpha$ Particle
1.0	0.56	0.64	0.63	1.81	1.92
2.0	0.92	1.23	1.21	3.15	3.39
3.0	1.18	1.50	1.63	4.12	4.52
4.0	1.42	1.82	2.03	4.94	5.47
5.0	1.62	2.11	2.41	5.68	6.31
6.0	1.82	2.37	2.75	6.34	7.07
7.0	1.98	2.61	3.03	6.92	7.76
8.0	2.15	2.84	3.31	7.52	8.43
9.0	2.31	3.05	3.55	8.08	9.07
10.0	2.45	3.24	3.80	8.64	9.67
15.0	3.11	4.10	4.83	10.8	12.3
20.0	3.68	4.88	5.72	13.0	14.5
25.0	4.20	5.58	6.51	15.2	16.9
30.0	4.66	6.21	7.33	17.0	19.1
35.0	5.08	6.81	8.02	18.7	20.7
40.0	5.49	7.34	8.72	20.1	22.2
45.0	5.87	7.89	9.33	21.3	23.4
50.0	6.24	8.38	9.95	22.4	24.7
60.0	6.93	9.32	11.02	24.4	27.2
70.0	7.55	10.2	12.0	26.5	29.8
80.0	8.06	11.0	13.1	28.4	32.1
90.0	8.72	11.7	14.0	30.6	34.4
100.0	9.25	12.5	14.8	32.4	36.6
150.0	11.61	15.7			
200.0	13.65	18.6			
300.0		23.3			
400.0		27.3			
500.0		30.7			



## APPENDIX 8

### ELEMENTARY PILE THEORY

Much of the material in this Appendix is based on a series of lectures in pile theory delivered by Dr. Harry Soodak as a part of the Clinton Laboratories Training School (1946-1947), of which school one of the authors was a member. A more rigorous treatment of this subject can be found in Soodak and Campbell, *Elementary Pile Theory*, John Wiley and Sons, 1950.

Here we propose to discuss in simple fashion some of the basic concepts of pile theory. For the sake of clarity we will limit our discussion to the case of thermal piles fueled by natural U or, at most, U slightly enriched in  $U^{235}$ . Such piles are sufficiently large that numerous simplifying approximations can be made. First we shall sketch the development of the so-called pile equations and show how these can be used to determine the critical dimensions for various pile shapes. Next we propose to investigate the effect of a reflector in reducing the critical size of the reactor.

#### Equations governing a finite pile

We assume here a pile composed of natural uranium spread in some suitable manner throughout a matrix of moderator. Although we know it is essential that the uranium be lumped, we shall ignore the local variations in neutron flux over the whole pile. This is not a bad assumption since the dips in thermal flux near U lumps produce only a minor local perturbation on the overall thermal neutron level. We further assume a pile of finite size, although its shape and dimensions are as yet unspecified.

If we wish to apply the four-factor formula of Chapter 11,

$$k_{\infty} = \eta \epsilon p f \quad (1)$$

to this pile, a number of changes are necessary. In the first place  $k_{\infty}$  must be replaced by  $k_{\text{eff}}$  since we are no longer presuming an infinite lattice. The quantities  $\eta$  and  $\epsilon$  are not affected by the change to a finite pile, but  $p$  and  $f$  are both disturbed. Hence for our finite pile we have instead of equation 1,

$$k_{\text{eff}} = \eta \epsilon p_{\text{eff}} f_{\text{eff}} \quad (1a)$$

We recall that  $p$  is the probability that a fast neutron with energy just below the  $U^{238}$  fission threshold reaches the thermal energy region. In addition to resonance capture by the uranium there now exists the opportunity for the neutron to "leak" from the pile, and hence  $p_{\text{eff}}$  will be less than  $p$ . Fermi and co-workers have shown that  $p_{\text{eff}}$  can be expressed by the following formula:

$$p_{\text{eff}} = p e^{-\tau K^2} \quad (2)$$

Here  $\tau$  is a quantity known as the neutron *age*. It is numerically equal to  $1/6$  the mean-square (crow-flight) distance from the point of origin of a neutron in the



pile to the point where it is finally reduced to thermal energy. In spite of the name,  $\tau$  does not in any way involve time. If one assumes suitable average values for  $\lambda_t$ , the transport mean free path\* of the neutron and  $\Sigma_s$ , the macroscopic scattering cross section, over the range from fission energy  $E_0$  to thermal energy  $E_{th}$ ,  $\tau$  can be calculated from the following expression:

$$\tau = \frac{\bar{\lambda}_t}{3} \left( \frac{\ln \frac{E_0}{E_{th}}}{\xi \Sigma_s} \right) \quad (3)$$

where  $\xi$ , applied to the moderator, has its usual meaning. Since  $\lambda_t$  and  $\Sigma_s$  vary with the neutron energy in a complicated manner it is usually preferable to determine  $\tau$  experimentally. For a typical U-graphite pile  $\tau$  has a value near 300 cm<sup>2</sup>. The quantity  $K^2$  is defined by the equation:

$$K^2 = - \frac{\nabla^2(n)}{n} \quad (4)$$

$n$  being the thermal neutron density within the pile ( $n$  the number of thermal neutrons per cubic centimeter), and  $\nabla^2$  representing the well-known Laplacian operator. In Cartesian coordinates

$$\nabla^2 = \frac{\delta^2}{\delta x^2} + \frac{\delta^2}{\delta y^2} + \frac{\delta^2}{\delta z^2} \quad (5)$$

Hence  $K^2$  can be written

$$K^2 = - \frac{\left( \frac{\delta^2 n}{\delta x^2} + \frac{\delta^2 n}{\delta y^2} + \frac{\delta^2 n}{\delta z^2} \right)}{n} \quad (6)$$

The proportionality of fast and thermal neutron density at every point in the pile is implicitly assumed in deriving the above expression for  $p_{eff}$ . This seems reasonable since slow neutrons are the agency for the generation of fast neutrons.

If one determines the thermal density along some line such as  $xx'$  or  $yy'$  of the pile depicted in Fig. 1, the curves shown will be obtained. The constant  $-K^2$  can be interpreted physically as measuring the curvature of the three-dimensional plot which represents the neutron distribution throughout the pile. As will be seen later, this constant is a very important property of any pile. Inasmuch as  $n$  in a going pile decreases as one proceeds from the center outward,  $K^2$  always will be positive for a chain-reacting system, and consequently  $p_{eff} < p$ .

\* If a material contains  $N$  atoms per cubic centimeter, each with a neutron scattering cross section of  $\sigma_s$ , the probability that a neutron moving in this material will be scattered in 1 cm of path is  $N\sigma_s \equiv \Sigma_s$ . It should be clear then that  $\lambda_s$ , the distance traveled between collision, is just  $1/\Sigma_s$ . Similarly,  $\lambda_a = 1/\Sigma_a$ . In scattering collisions by neutrons with light nuclei, forward scattering is slightly favored. Hence, in a given number of collisions the neutron will actually progress farther than it would if the scattering were spherically symmetrical. To take account of this effect one introduces the transport mean free path  $\lambda_t$ . It turns out that  $\lambda_t = \lambda_s/[1 - (2/3A)]$  where  $A$  is the mass number of the scattering nucleus. Obviously, for large  $A$ ,  $\lambda_t \approx \lambda_s$ .



$f$ , the fraction of thermal neutrons absorbed by uranium, is also here decreased by the escape of some through the surface of our finite pile. It turns out that  $f_{\text{eff}}$  is given by

$$f_{\text{eff}} = \frac{f}{(1 + L_p^2 K^2)} \quad (7)$$

where  $K^2$  has the same meaning as before, and  $L_p$  is the diffusion length for thermal neutrons in the pile.  $L_p^2$  is given numerically by  $\frac{1}{6}$  the mean-square (crow-flight) distance from the point where the neutron becomes thermal to the point in the pile where it is finally absorbed.  $L_p^2$  is defined by the following equation:

$$L_p^2 = \frac{\lambda_a \lambda_t}{3} \quad (8)$$

where  $\lambda_t$  is the transport mean free path for a thermal neutron, and  $\lambda_a$  is the mean free path for absorption in the pile. Because it is most difficult to calculate  $\lambda_a$  for a heterogeneous mixture of uranium and moderator one usually proceeds as follows to deduce  $L_p$ :

1. Determine by experiment the diffusion length  $L_m$  for thermal neutrons in the moderating medium alone.

2. It has been shown by Plass that to a first approximation

$$L_p^2 = L_m^2(1 - f) \quad (9)$$

3. If the uranium and moderator were uniformly dispersed,  $f$  would be simply

$$f = \frac{N_u \sigma_u}{N_m \sigma_m + N_u \sigma_u} \quad (9a)$$

where  $N_u$  and  $N_m$  represent the number of uranium and moderator atoms per cubic centimeter of pile, and  $\sigma_u$  and  $\sigma_m$  are their respective absorption cross sections. For a lumped lattice the situation is much more complicated.

However, from a knowledge of the size and spacing of the uranium lumps in the pile and the nuclear constants involved, it is possible to work out an expression for  $f$ . This is then substituted in equation 9 to give  $L_p^2$ . For a typical U-graphite pile  $L_p^2$  also has a value near 300 cm<sup>2</sup>.

Now the condition that a pile maintain a chain reaction is just

$$k_{\text{eff}} = 1 \quad (10)$$

Therefore, combining equations 1a, 2, 7, and 10, we have

$$\eta \epsilon p f e^{-\tau K^2} \left( \frac{1}{1 + L_p^2 K^2} \right) = 1 \quad (11)$$

But, since  $k_{\infty} = \eta \epsilon p f$ , equation 11 can be written

$$k_{\infty} - (1 + L_p^2 K^2) e^{\tau K^2} = 0 \quad (12)$$

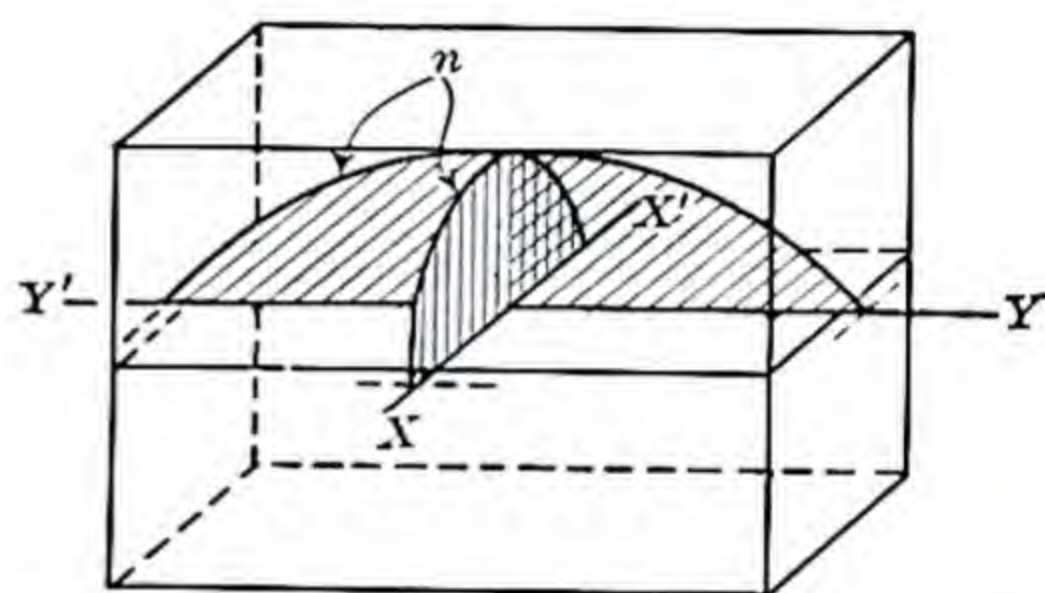
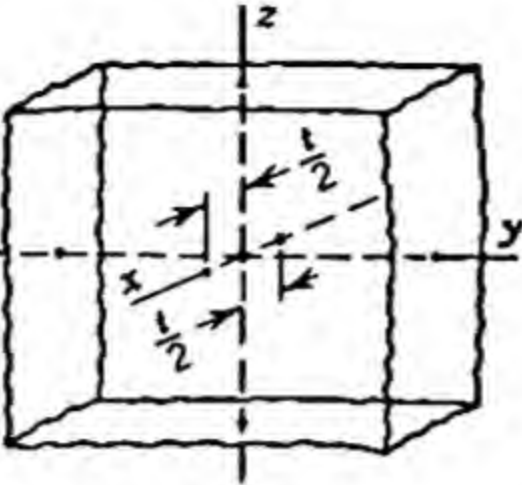
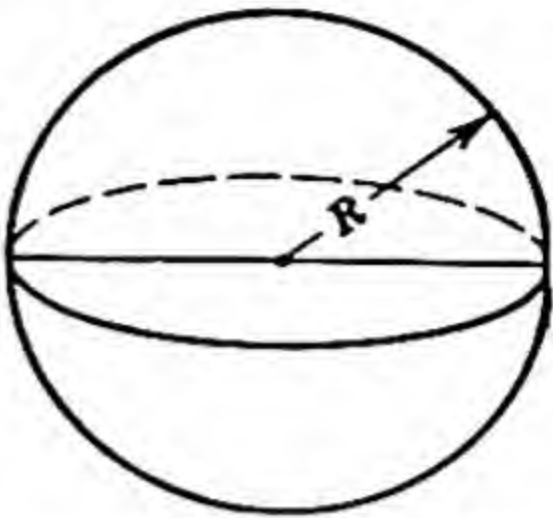


FIG. 1. Rectangular block pile, showing the manner in which the neutron density varies along  $xx'$  and  $yy'$ . Note in particular that the density goes to zero at a point beyond the physical boundary of the block.

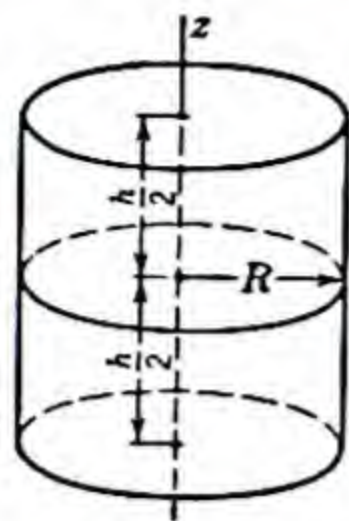


TABLE

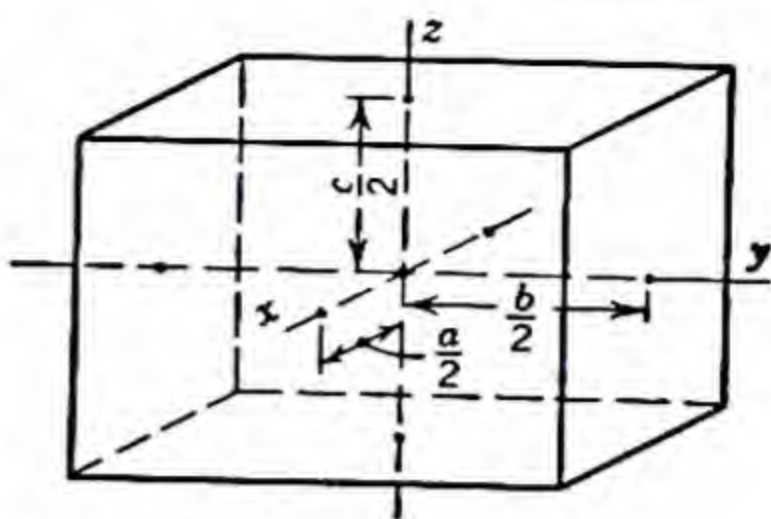
Sketch of pile		
Type of pile	Slab pile infinite in $y$ and $z$ directions	Spherical pile
Boundary of pile	Planes $x = \pm \frac{l}{2}$	Sphere of radius $R$
Equation 4 *	$\frac{d^2(n)}{dx^2} + K^2n = 0$	$\frac{d^2(n)}{dr^2} + \frac{2}{r} \frac{d(n)}{dr} + K^2n = 0$
Solution of equation 4 for $K^2 > 0$ , i.e., $K$ must be real †	$n = A \cos Kx$	$n = \frac{A}{r} \sin Kr$
Conditions for $n = 0$ at boundary §	$K = \frac{\pi}{l}$ $\therefore K^2 = \frac{\pi^2}{l^2}$	$K = \frac{\pi}{R}$ $\therefore K^2 = \frac{\pi^2}{R^2}$
Critical dimensions	$l = \frac{\pi}{K}$	$R = \frac{\pi}{K}$
Critical volume	$V = \infty$	$V = \frac{4}{3}\pi R^3$ $V = \frac{130}{K^3}$



1



Cylindrical pile



Rectangular block pile

Circular cylinder of radius  $R$   
planes  $z = \pm \frac{h}{2}$

Planes  
 $x = \pm \frac{a}{2}$   
 $y = \pm \frac{b}{2}$   
 $z = \pm \frac{c}{2}$

$$\frac{\delta^2(n)}{\delta r^2} + \frac{1}{r} \frac{\delta(n)}{\delta r} + \frac{\delta^2(n)}{\delta z^2} + K^2 n = 0$$

$$\frac{\delta^2(n)}{\delta x^2} + \frac{\delta^2(n)}{\delta y^2} + \frac{\delta^2(n)}{\delta z^2} + K^2 n = 0$$

$$n = A(\cos K_1 z)(J_0[K_2 r])$$

where  $K^2 = K_1^2 + K_2^2$  ‡

$$n = A \cos(K_1 x) \cos(K_2 y) \cos(K_3 z)$$

where  $K^2 = K_1^2 + K_2^2 + K_3^2$

$$K_1 = \frac{\pi}{h} \quad K_2 = \frac{2.405}{R}$$

$$\therefore K^2 = \frac{\pi^2}{h^2} + \frac{(2.405)^2}{R^2}$$

$$K_1 = \frac{\pi}{a} \quad K_2 = \frac{\pi}{b} \quad K_3 = \frac{\pi}{c}$$

$$\therefore K^2 = \frac{\pi^2}{a^2} + \frac{\pi^2}{b^2} + \frac{\pi^2}{c^2}$$

For minimum critical volume  
 $R = 0.541h$

For minimum critical volume

$$\text{Thus: } h = \frac{5.441}{K}; R = \frac{2.945}{K} \parallel$$

$$\text{Cube: } a = b = c = \frac{\sqrt{3}\pi}{K} = \frac{5.44}{K}$$

Minimum critical volume

$$V = \pi R^2 h$$

$$V = \frac{148.2}{K^3}$$

Minimum critical volume

$$V = a^3$$

$$V = \frac{161}{K^3}$$

(Reference notes for table are on following page)



Table 2 lists the critical dimensions for the various pile shapes, utilizing the above value of  $K^2$ . As might be expected, the spherical pile boasts the smallest volume, followed by the cylindrical pile and cubical pile in that order.

TABLE 2

PILE SHAPE	CRITICAL DIMENSION	CRITICAL DIMENSION CORRECTED FOR AUGMENTED DISTANCE	CRITICAL VOLUME
Slab	$l = 314 \text{ cm}$	$l = 310 \text{ cm}$	$V = \infty$
Spherical	$R = 314 \text{ cm}$	$R = 312 \text{ cm}$	$V = 1.27 \times 10^8 \text{ cm}^3$
Cylindrical	$R = 294.5 \text{ cm}$ $h = 544 \text{ cm}$	$R = 292.5 \text{ cm}$ $h = 540 \text{ cm}$	$V = 1.48 \times 10^8 \text{ cm}^3$
Cubical	$a = 544 \text{ cm}$	$a = 540 \text{ cm}$	$V = 1.58 \times 10^8 \text{ cm}^3$

The three functions that describe the variation of neutron density within our four piles are:

$$(1) \cos \frac{\pi}{2} \alpha \quad \text{where } \alpha = \frac{2x}{l} \text{ for slab or } \frac{2z}{h} \text{ for cylinder}$$

$$(2) \frac{\sin \pi \alpha}{\pi \alpha} \quad \text{where } \alpha = \frac{r}{R} \text{ for sphere}$$

$$(3) J_0(2.405\alpha) \quad \text{where } \alpha = \frac{r}{R} \text{ for cylinder}$$

These functions are plotted in Fig. 2 over the range  $\alpha = 0$  (center of pile) to 1 (pile boundary). It is clear that they differ only slightly in form. Hence as one moves out from the center of a pile along a major axis no appreciable error is made in assuming that the neutron density follows a cosine function in all cases.

#### Pile with reflector

Thus far we have limited our discussion to bare piles. In practice a reflector almost invariably surrounds the reactor. By adding a reflector one secures two major advantages.

1. The critical size of the reactor is appreciably reduced. This permits a considerable saving in the amount of fissionable material needed for a chain-reacting pile.

2. The neutron distribution over the pile shows less variation than in the bare pile case (see Fig. 3). This means that the fuel near the reactor boundary is entering more fully into the chain reaction. Thus if the power level is limited by the temperature of fuel rods at the center, a pile with a reflector can be operated at a higher power output than the same pile without a reflector. It is assumed of course that the latter will be chain reacting. The chief purpose of the reflector is to scatter back into the pile where they can be utilized neutrons which would otherwise leak and be lost. Therefore in selecting a reflector medium one seeks a material with a large  $\Sigma_s$  for neutrons and low  $\Sigma_a$ . For these reasons good moderators usually make good reflectors. To date graphite has proved most popular for reflector purposes, just as it has been the preferred moderator. However, one should not exclude Be,



BeO, D<sub>2</sub>O, and even H<sub>2</sub>O. In fact, water in small thicknesses is a very excellent reflector because of its high  $\Sigma_s$ . For thicker layers the relatively large  $\Sigma_a$  of water predominates to make it less attractive.

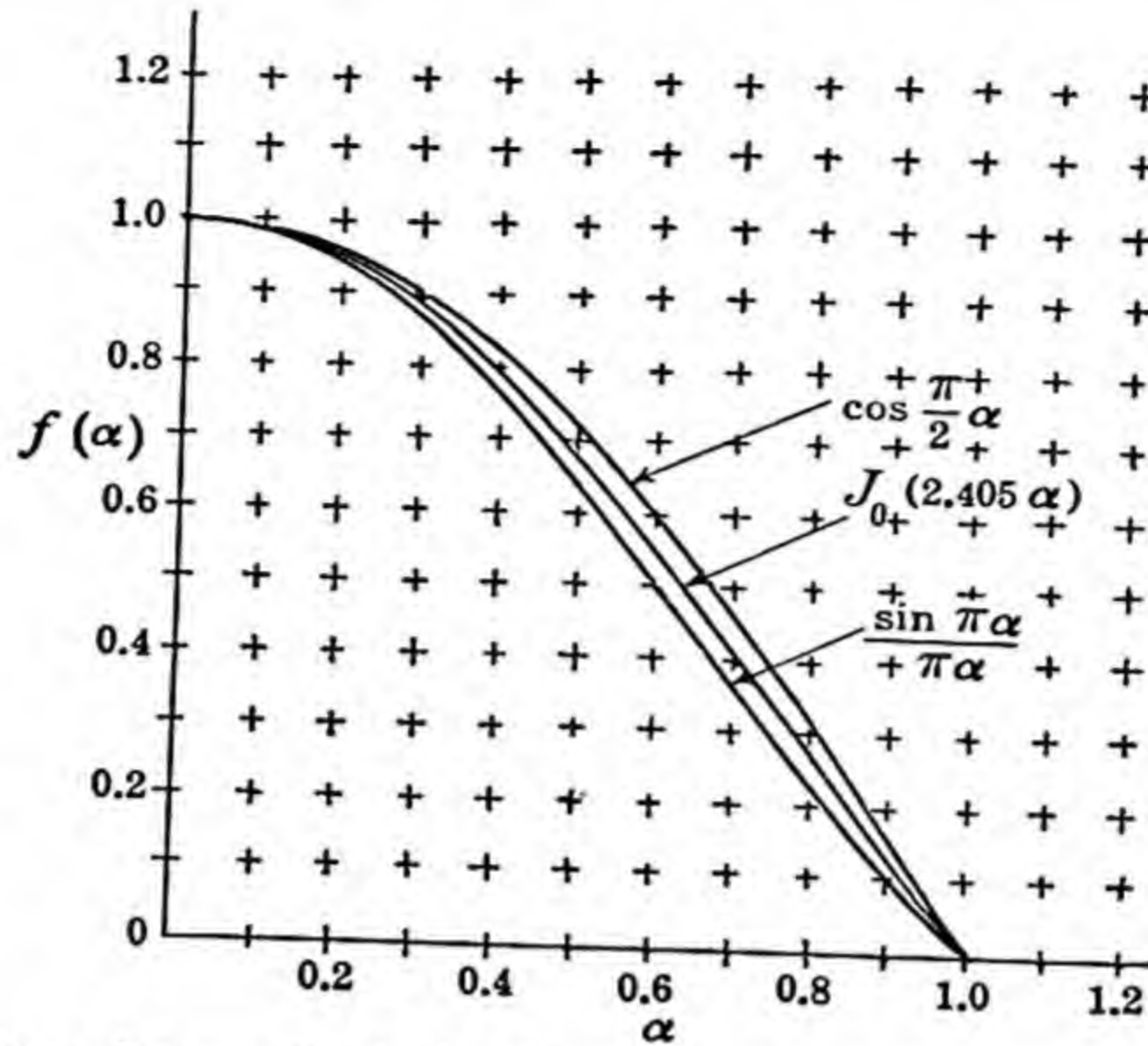


FIG. 2. Plot showing the marked similarity in behavior of the functions used to express the neutron density within a chain reactor.

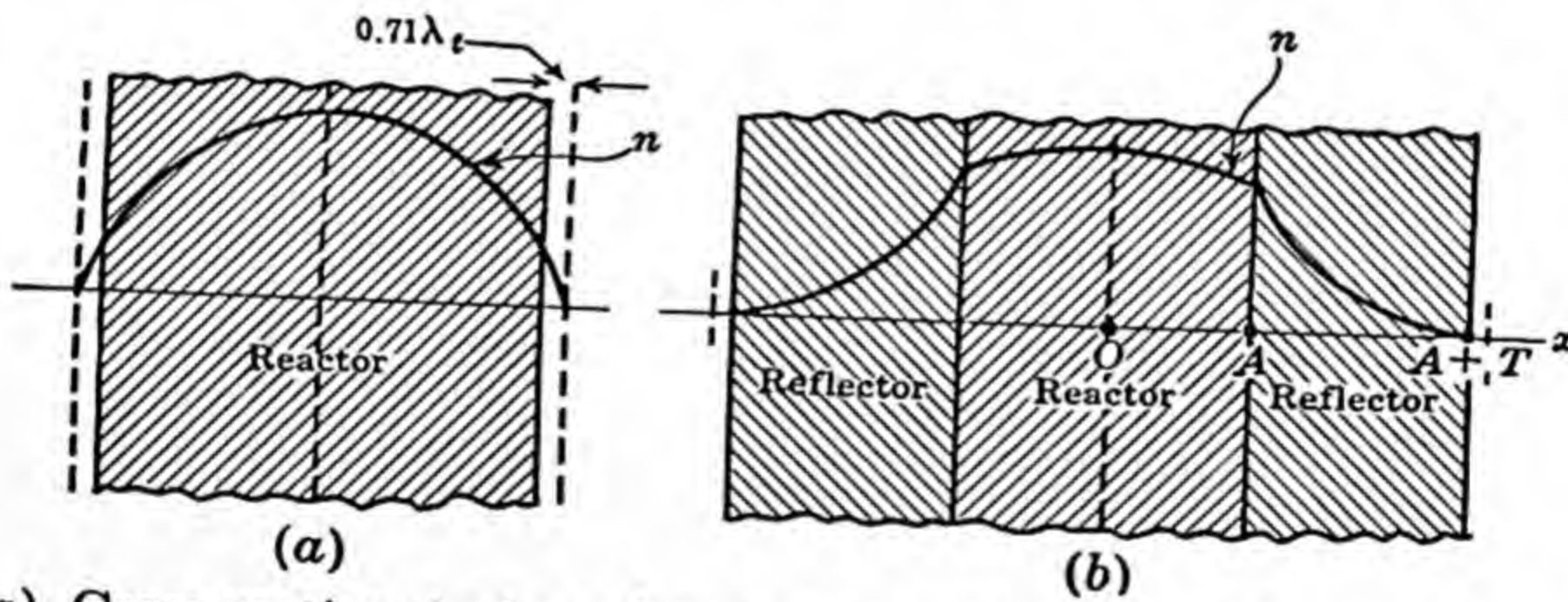


FIG. 3. (a) Cross-sectional view showing a portion of a bare slab pile and the density distribution across the pile. (b) View of a slab pile with reflector added. Note the decreased reactor thickness as compared with (a); also the flattened neutron distribution within the reactor. In the reflector, since no multiplication occurs, neutron density rapidly decreases with distance. The rate of decrease is determined by the reflector thickness and the diffusion length  $L$  for neutrons in the reflector.

As an illustration of how effective a reflector can be in reducing the critical size of a reactor we shall solve this problem for the slab pile shown in cross section in Fig. 3b. In order to reduce the complexity of the calculation we will make the following oversimplifying assumptions:

- (1) All neutrons present have the same energy (thermal).
- (2) Moderator and reflector are composed of the same material (graphite).

The first assumption ignores the fact that fast neutrons are present both in the reactor and reflector. It leads us to underestimate slightly the effectiveness of the



reflector since the latter possesses a higher albedo (reflection coefficient) for fast than for slow neutrons. Once the proposed problem is solved we will indicate what changes are necessary when condition 2 is not satisfied.

To start, let us combine the pile equations 4 and 13 into a form which will hold (1) in the reactor and (2) in the reflector. Since no slowing down is involved under our simple approximation,  $\tau$  will be everywhere zero. Therefore in the reactor we find

$$\nabla^2(n) + \frac{k_\infty - 1}{L_p^2} n = 0 \quad (14)$$

where we have substituted the value of  $K^2$  from equation 13 into equation 4.

In the reflector  $k_\infty = 0$ . Hence the equation applicable to this region is

$$\nabla^2(n) - \frac{1}{L_r^2} n = 0 \quad (15)$$

where  $L_r$  is the thermal diffusion length in the reflector, i.e., in pure graphite.

We must find solutions for equations 14 and 15 subject to the following boundary conditions (see Fig. 3b).

- (1)  $n \neq 0$  at the origin ( $x = 0$ ).
- (2)  $n = 0$  at the outer boundary of the reflector ( $x = A + T$ ).
- (3)  $n$  and  $dn/dx$  be continuous at the reactor-reflector interface, ( $x = A$ ).

Solutions that satisfy conditions 1 and 2 are

$$n = C_1 \cos \frac{\sqrt{k_\infty - 1}}{L_p} x \quad \text{for } x \leq A \quad (16)$$

$$n = C_2 \sinh \left[ \frac{1}{L_r} (T + A - x) \right] \quad \text{for } A \leq x \leq A + T \quad (17)$$

$C_1$  and  $C_2$  are constants.

The first part of condition 3 requires that

$$C_1 \cos \phi_1 A = C_2 \sinh \phi_2 T \quad (18)$$

where we have substituted

$$\phi_1 = \frac{\sqrt{k_\infty - 1}}{L_p} \quad \text{and} \quad \phi_2 = \frac{1}{L_r}$$

The second part of 3 demands that

$$-C_1 \phi_1 \sin \phi_1 A = -C_2 \phi_2 \cosh \phi_2 T \quad (19)$$

Dividing equation 19 by equation 18 yields

$$\phi_1 \tan \phi_1 A = \phi_2 \coth \phi_2 T \quad (20)$$

Now as  $T \rightarrow 0$ ,  $\coth \phi_2 T \rightarrow \infty$ . This requires that the argument of the  $\tan \rightarrow \pi/2$ , i.e.,  $\phi_1 A_0 = \pi/2$ , or

$$A_0 = \frac{\pi}{2\phi_1} \quad (21)$$



Here  $A_0$  is just the critical half-thickness of the slab pile with no reflector.

The critical value of  $A$  with a reflector present is, from equation 20,

$$A = \frac{1}{\phi_1} \tan^{-1} \left( \frac{\phi_2}{\phi_1} \coth \phi_2 T \right) \quad (22)$$

The reduction in half-thickness of the reactor due to the reflector is just

$$S \equiv A_0 - A \quad (23)$$

which is the quantity we desire to calculate.  $S$  is known by the descriptive title "pile savings."

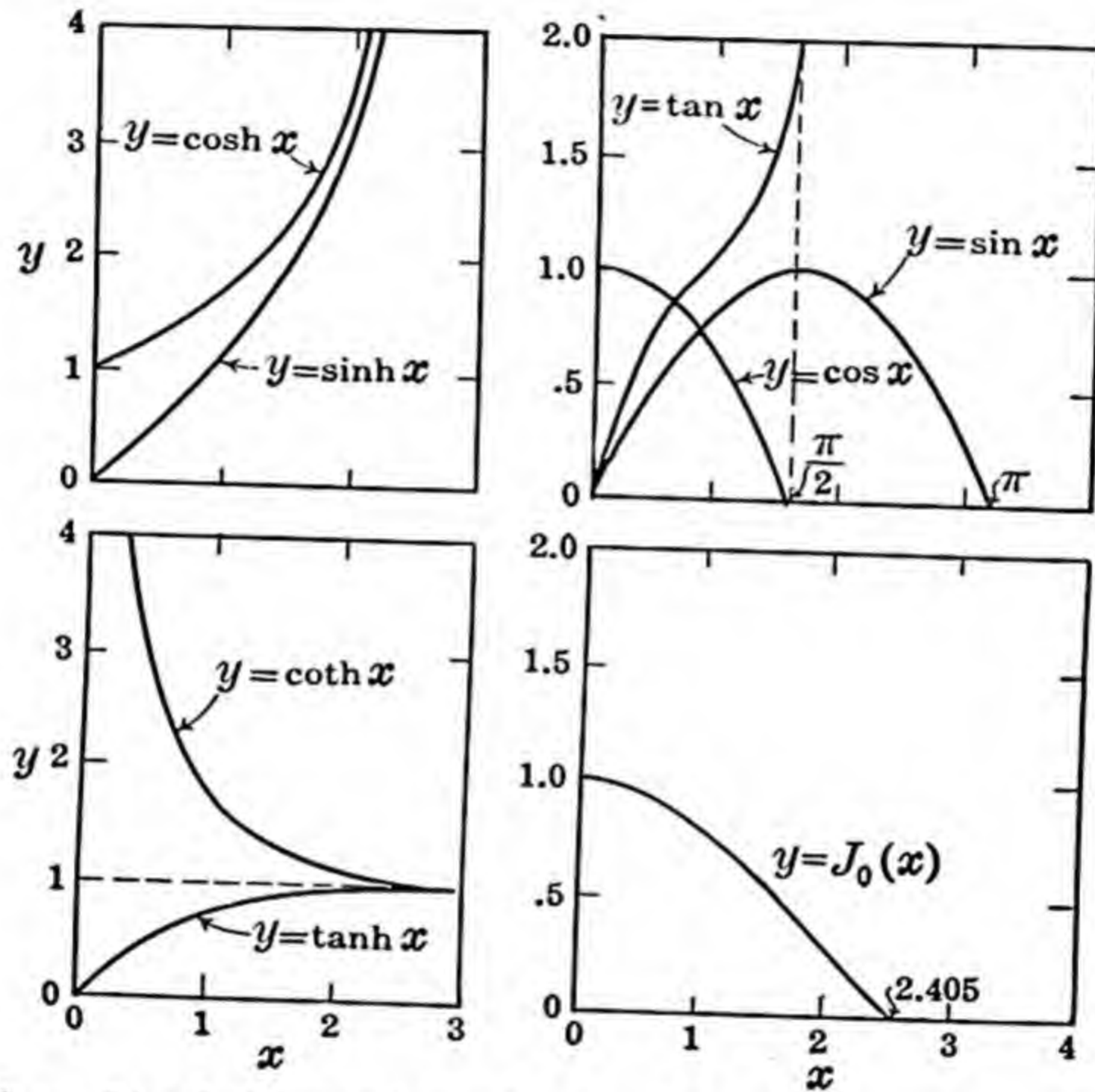


FIG. 4. For the benefit of the reader who is unfamiliar with the various functions involved in solving the pile equations we include the above plots. We record their behavior in the first quadrant alone (positive values of  $x$  and  $y$ ) since this is the region of interest for steady-state pile applications. Both  $x$  and  $y$ , the values of the argument and the function, respectively, are dimensionless quantities.

Substituting equations 21 and 22 into equation 23 gives:

$$S = \frac{1}{\phi_1} \left[ \frac{\pi}{2} - \tan^{-1} \left( \frac{\phi_2}{\phi_1} \coth \phi_2 T \right) \right] \quad (24)$$

In evaluating  $\phi_1$  and  $\phi_2$  we will once more use values published by Fermi for a uranium-graphite lattice, namely,  $k_\infty = 1.06$ ,  $L_p = 17.4$  cm, and  $L_r = 50$  cm (pure graphite). These lead to  $\phi_1 = 0.014$  cm $^{-1}$  and  $\phi_2 = 0.020$  cm $^{-1}$ .

With these values substituted in equation 24 one is ready to find the pile savings for any reflector thickness. Table 3 tabulates  $S$  for various values of  $T$ .



TABLE 3

PILE SAVINGS  $S$  FOR A SLAB PILE, ASSUMING VARIOUS THICKNESSES OF REFLECTOR (GRAPHITE)

$T$ (cm)	$S$ (cm)
1	1
5	5
10	10
50	35
100	42.2
$\infty$	43.2

It is seen that a thin layer of graphite ( $\sim 10$  cm) reduces the reactor half-thickness by a corresponding amount. On the other hand little is gained in making a graphite reflector thicker than 100 cm.

It is interesting to calculate the reduction in size for the cubic pile of Table 2 if we surround the reactor with a 1-meter layer of graphite. Strictly speaking, the value of 42.2 cm found for this case applies only to a slab pile. However, owing to the large size of a critical natural uranium-graphite cube, we can apply the same correction without introducing an appreciable error. The new value for the cube edge will be  $540 - 84 = 356$  cm. This gives a critical volume of only  $0.45 \times 10^8$  cm<sup>3</sup>, compared to  $1.58 \times 10^8$  cm<sup>3</sup> for a bare pile. The surprising conclusion is that by the addition of a 1-meter graphite reflector we require only 28.6 per cent as much uranium to construct a chain-reacting system as would be needed in the absence of a reflector.

If the moderator and reflector differ in composition equation 24 must be replaced by

$$S = \frac{1}{\phi_1} \left[ \frac{\pi}{2} - \tan^{-1} \left( \frac{\lambda_{t2}\phi_2}{\lambda_{t1}\phi_1} \coth \phi_2 T \right) \right] \quad (25)$$

where  $\lambda_{t2}$  is the transport mean free path for a neutron in the reflector, and  $\lambda_{t1}$  is the same quantity for the moderator. If we apply equation 25 to the uranium-graphite lattice considered above but employ an H<sub>2</sub>O reflector we must use the following constants, again gleaned from a publication by Fermi:

$$\begin{aligned} k_{\infty} &= 1.06 & \phi_1 &= 0.014 \text{ cm}^{-1} & \lambda_{t1} &= 2.7 \text{ cm (graphite)} \\ L_p &= 17.4 \text{ cm} & \phi_2 &= 0.35 \text{ cm}^{-1} & \lambda_{t2} &= 0.43 \text{ cm (water)} \\ L_r &= 2.85 \text{ cm (pure water)} \end{aligned}$$

Table 4 lists the pile savings for this case. This table bears out the previous statement that a thin layer of H<sub>2</sub>O makes an excellent reflector. But owing to the large value of  $\phi_2$  ( $\phi_2 \propto \Sigma_a$ ) an infinite thickness of water is much less effective than an infinite graphite layer.

It must be emphasized that all reactor dimensions calculated in this chapter refer to piles with a maximum  $k_{eff}$  of 1, i.e., they will just be chain reacting. In practice a pile is usually built larger than the critical dimensions, the excess reactivity being controlled by means of regulating rods (thermal piles) or gap controls (resonance and fast piles). The extra reactivity present provides a potential excess of neutrons for



experimental purposes and also allows one to overcome the decrease in  $k$  due to fuel depletion, fission product poisoning, and changes in temperature.

TABLE 4

PILE SAVINGS  $S$  FOR A SLAB PILE, ASSUMING VARIOUS THICKNESSES OF REFLECTOR ( $\text{H}_2\text{O}$ )

$T$ (cm)	$S$ (cm)
1	6
5	16.4
10	17.5
50	17.5
$\infty$	17.5

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- C. GOODMAN (editor), *The Science and Engineering of Nuclear Power*, Addison-Wesley Press, Cambridge, Mass., Vol. I, 1947, Chapters 3, 5, and 6; Vol. II, 1949, Chapter 4.
- H. SOODAK and E. C. CAMPBELL, *Elementary Pile Theory*, John Wiley and Sons, Inc., 1950.



## APPENDIX 9

### NEUTRON DIFFRACTION

According to de Broglie any particle of mass  $m$  moving with velocity  $v$  has an associated wavelength  $\lambda$ .

$$\lambda = \frac{h}{mv} = \frac{h}{\sqrt{2mE}}$$

$h$  = Planck's constant, and  $E$  = kinetic energy of the particle. By the use of this formula one finds that thermal neutrons possess wavelengths of the same order of magnitude as x-rays employed in diffraction work. This immediately

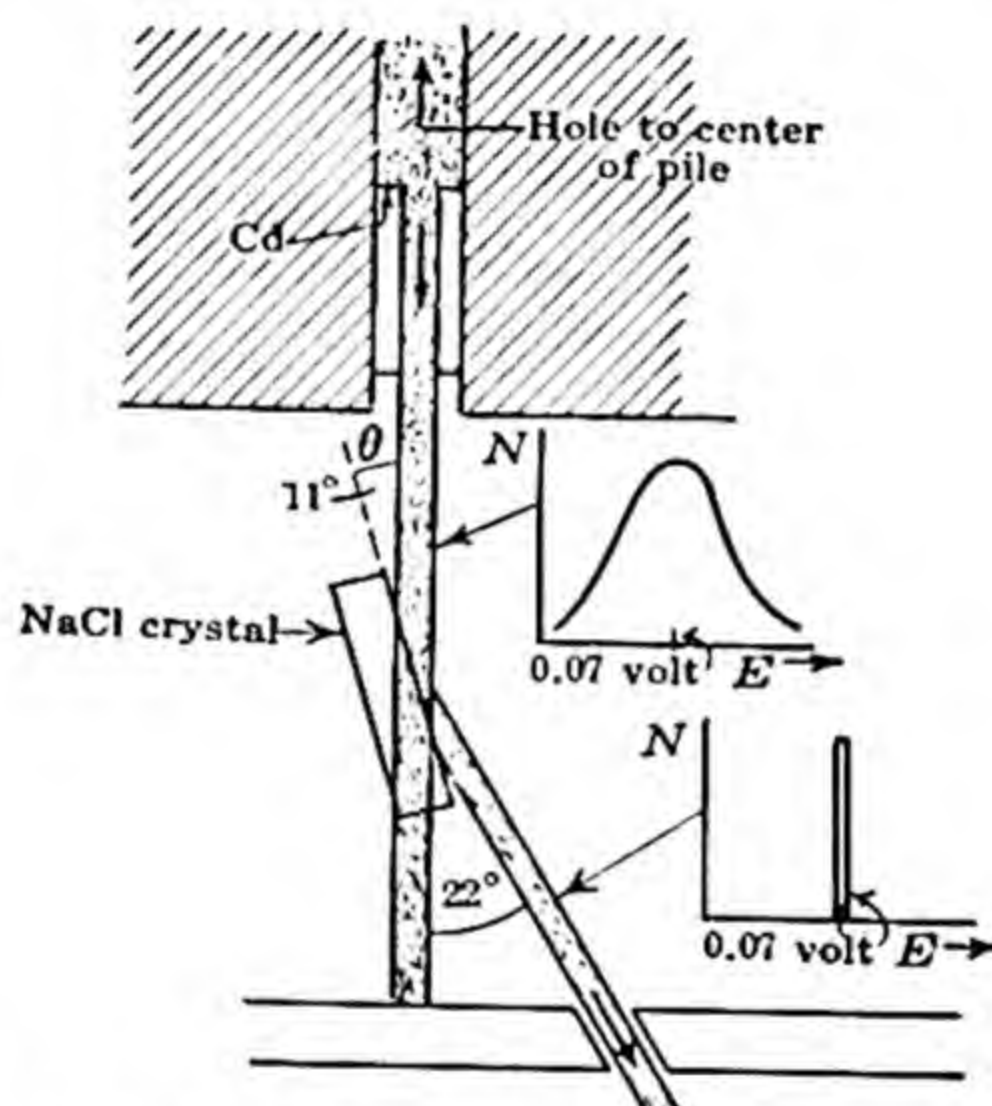


FIG. 1. Diagram shows principle of the neutron spectrometer. A single crystal (NaCl, LiF, etc.) intercepts the collimated neutron beam at an angle  $\theta$  such that a prominent set of crystal planes is in diffracting position for neutrons of the energy desired. For 0.07-volt neutron and the 200 plane in NaCl the required value of  $\theta$  is 11 degrees. The insert diagrams picture the energy distribution in the incident and diffracted neutron beams, the latter being essentially monochromatic. Various valuable diffraction and absorption experiments can be performed using this beam.

arouses speculation as to the possibility of using slow neutron beams as a diffraction tool. Even though this possibility was recognized many years ago it was of mere academic interest, owing to the weak intensity and spread-out energy of thermal neutron beams obtainable at that time. The advent of chain-reacting piles with their high neutron fluxes placed this matter in a new



light, and the technique of neutron diffraction has already been applied to a variety of problems. Admittedly the thermal neutrons issuing from a pile show a broad Maxwellian spread in energy, but Fig. 1 illustrates how a "monochromatic" beam of neutrons may be secured by diffraction from a single crys-

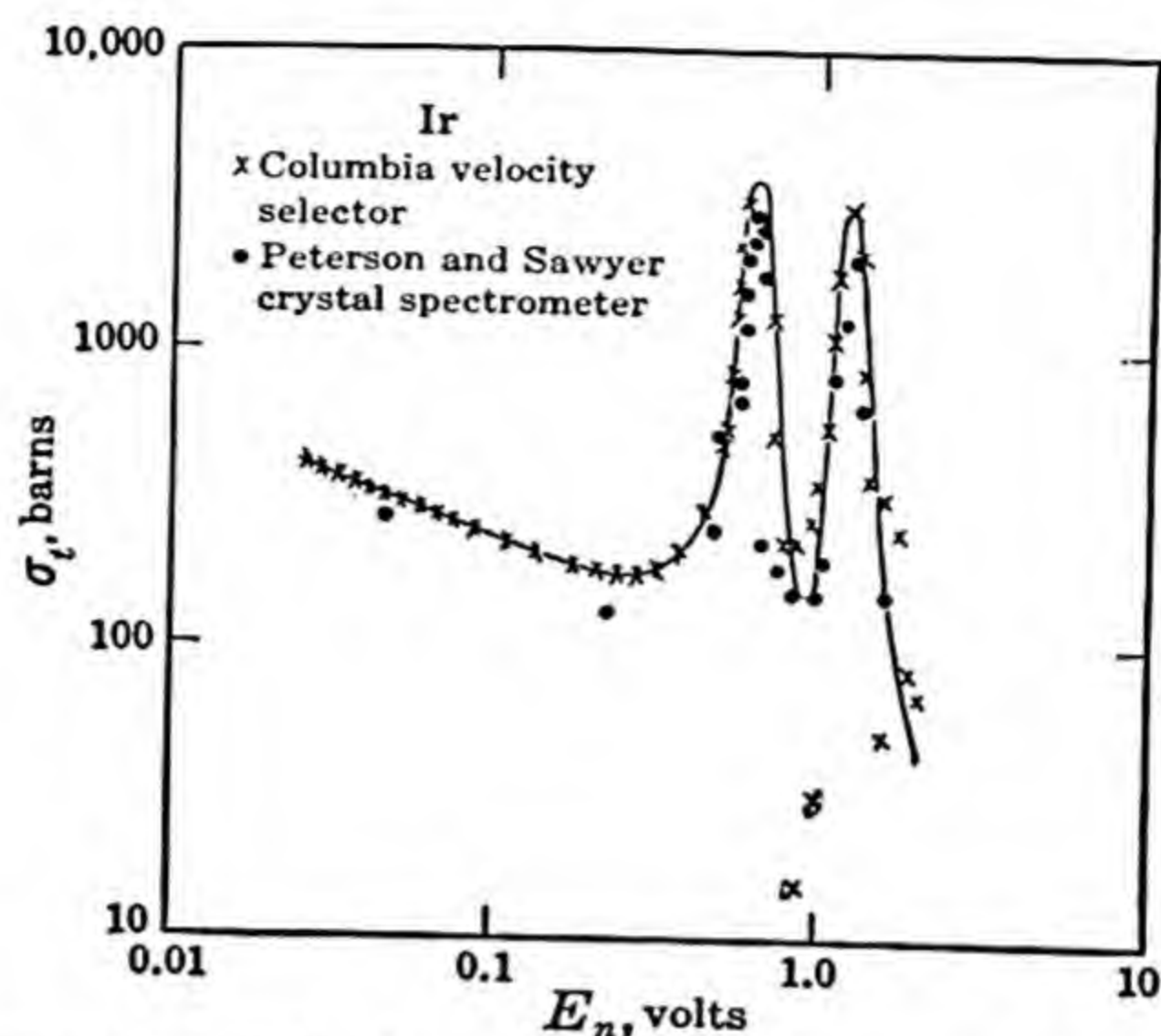


FIG. 2. Plot showing the variation in  $\sigma_t$  with energy for Ir over the thermal neutron energy region. Notice the close agreement between values obtained by the "time of flight" and neutron spectrometer methods. In addition to  $\sigma_t$  the spectrometer method is capable of measuring  $\sigma_a$  as a function of energy by means of activation measurements.

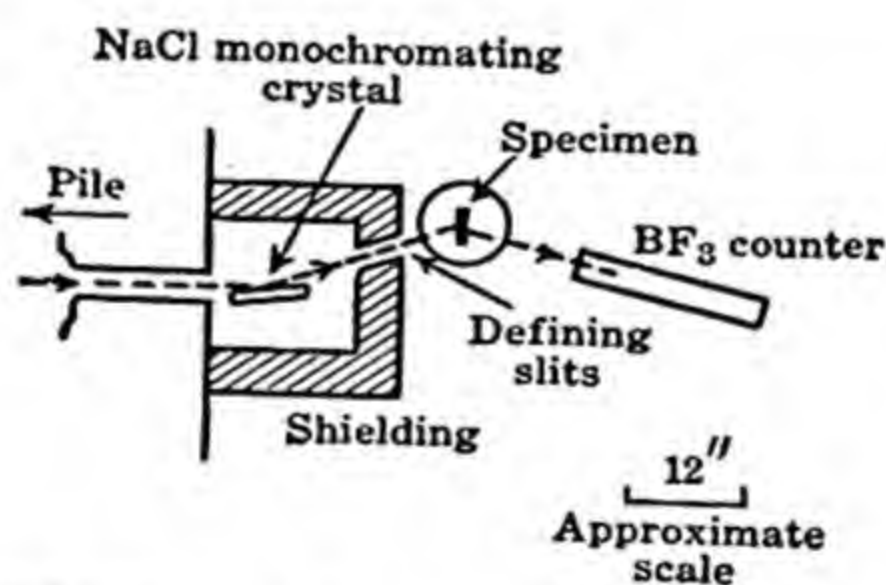


FIG. 3. Sketch of the neutron spectrometer used by Wollan and Shull at Oak Ridge for determining the phase of neutron scattering in elements and for crystal structure studies. Neutrons diffracted in the powder specimen are detected by the long  $\text{BF}_3$  counter which can be rotated about an axis concentric with that of the specimen table. In this manner the powder diffraction pattern is recorded. Auxiliary equipment permits automatic traversal and recording of any desired angular interval.

tal. By changing the angle between the incident beam direction and crystal face a different segment of the neutron distribution is diffracted. This apparatus is termed a "neutron spectrometer" and with it one can produce at will monochromatic neutron beams over a considerable range of energy in the fractional electron-volt region. This scheme has been used extensively in measuring



the total neutron cross section for elements in the interval from 0.05 to 5 electron volts, where it supplements the "time of flight" method very nicely. Figure 2 depicts the results obtained on Ir by both methods. Satisfactory agreement is to be noted. Since the spectrometer provides spatial separation of



FIG. 4. Neutron diffraction by a rock-salt crystal, as recorded by Wollan and Shull of the Oak Ridge National Laboratory.

neutron energies one has a means of determining absorption cross sections uniquely through measurements on the induced activities.

From a record of the diffraction pattern from a specimen, using the setup sketched in Fig. 3, it is possible to evaluate the "phase" of neutron scattering for various elements and the method is much used in this connection. However, potentially the most powerful application of neutron diffraction is in the field of crystal structure analysis. For a long time x-ray diffraction has exer-



cised a monopoly in this field, but there are many places where x-rays prove inadequate. For the study of superstructure in alloys of metals with neighboring  $Z$  values (Fe and Co, for example) x-rays offer little help, owing to the close similarity in x-ray scattering power for these two atoms. Likewise it has previously been difficult or impossible to pinpoint hydrogen atom positions in H-containing crystals on account of the negligible scattering of x-rays by the lone electron associated with hydrogen. Fortunately neutron diffraction does not suffer this limitation. Thermal neutron-scattering cross sections conform to no set pattern as regards atomic number, hydrogen having an outstandingly high value. Similarly Fe and Co appear decidedly different to thermal neutrons. Scattering cross section is not a completely adequate criterion here, since only the coherent part of the scattering acts to produce a diffraction pattern. Nevertheless it has been possible to identify superstructure formation in Fe-Co alloys via neutron diffraction, to deduce "magnetic" unit cells in ferromagnetic and antiferromagnetic crystals, and the method has been used successfully to locate hydrogen positions in the crystals NaH and H<sub>2</sub>O (ice). A most promising future is predicted for this technique once the number of nuclear reactors for research purposes is increased to the point where the tool will be available to crystallographers on a routine basis.

As an example of neutron diffraction we show in Fig. 4 the pattern produced by a single crystal of sodium chloride. The bright patch at the center is due to undiffracted neutrons; the pattern of spots are at angles for which the de Broglie wavelength is right for diffraction. Any reader who may not be convinced of the wave nature of matter should look at this picture and recall that the neutron is an uncharged particle.

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- E. O. WOLLAN, W. L. DAVIDSON, and C. G. SHULL, "Neutron Diffraction Study of the Structure of Ice," *Phys. Rev.*, **75**, 1348 (1949).
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## APPENDIX 10

### CROSS SECTIONS

For fast particles Serber has developed a very interesting viewpoint. The incoming particle is regarded as a wave which encounters regions of different refractive index and finite size. These regions are the nuclei. For light nuclei the absorption is not complete so that a neutron has about a 50 per cent chance of going through the nucleus. Apart from this the chance of absorption is given by a nuclear cross section  $\pi r^2$  where  $r = 1.37 \times 10^{-13} A^{1/3}$ . For heavy elements the chance of being retained is unity. The scattering cross section is larger, being determined by the diffraction of the wave. For charged particles the coulomb barrier complicates this, though only appreciably so, at less than 10 Mev.

For slow particles, which are necessarily neutrons, the cross section  $\sigma$  is dominated by the resonance formula

$$\sigma = \sigma_0 \frac{\Gamma^2}{(E - E_0)^2 + \frac{\Gamma^2}{4}}$$

Superposed on this is the non-resonant cross section varying as  $1/v$ . The entire description of the cross section is often rather more complicated, and a complete curve for  $\sigma$  versus energy must be obtained experimentally. The available material is admirably summarized by Goldsmith, Ibser, and Feld in a table published by the Addison Wesley Press, Cambridge, Massachusetts. This table should be consulted for any detailed knowledge. We give below a selection of cross sections chosen more from the point of view of interest than for reference.



INCIDENT PARTICLE	ELE- MENT	NATURE OF CROSS SECTION	LAW OBEYED	ENERGY (elec- tron volts)	CROSS SECTION (barns)	RESO- NANT ENERGY (elec- tron volts)	CROSS SECTION AT RESO- NANCE (barns)	HALF- WIDTH AT RESO- NANCE
n	H	Total	Not simple	0.1	36			
		Scattering	Not simple	0.01	71			
	D	Total	Not simple	0.1	13			
				1.0	4			
	Li	Total	Not simple	0.1	3			
				1.0	1			
	B	Total	1/v	0.1	40			
	Al	(n $\alpha$ )	1/v	0.1	350			
		Total	Roughly constant	1.0	1.5			
	Mn	Total	1/v plus reso- nance	0.1	10	300	18	1,000
	Rh	Total	1/v plus reso- nance	0.1	90	1.3	2,000	0.3
	Ag	Total	1/v plus reso- nance	0.1	35	5.2	500	1.2
						16.0	22	3
	Cd	Total	1/v plus reso- nance	0.01	3200	45	16	18
						0.176	7,200	0.13
	Sm	Total	Complex reso- nance			0.1	15,000	0.07
	Gd	Total	1/v falling off	0.01	$7 \times 10^4$			
	Elec- tron	Total	Not established	0.1	$10^{-13}$			
<hr/>								
$\gamma$	D	Nuclear photo effect		2.5 (Mev)	0.001			
e	Rh	Electron excitation		0.8 (Mev)	$10^{-8}$			
Proton	C	Formation of $\pi$ -mesons	Partial expo- nential rise	350 (Mev)	0.01			
Alpha particle	C	Formation of $\pi$ -mesons	Partial expo- nential rise	350 (Mev)	0.001			



## APPENDIX 11

### SOME LABORATORY EXPERIMENTS

In order to bring some reality to one's understanding of nuclear physics, a few chances to manipulate equipment and see what it does are invaluable. Such experiments are now relatively cheap and easy, and we wish to suggest briefly a few that can be tried.

The nuclear part of the equipment, the radioactive sources, can readily be obtained at tolerable cost. We suggest a few millicuries of polonium as a

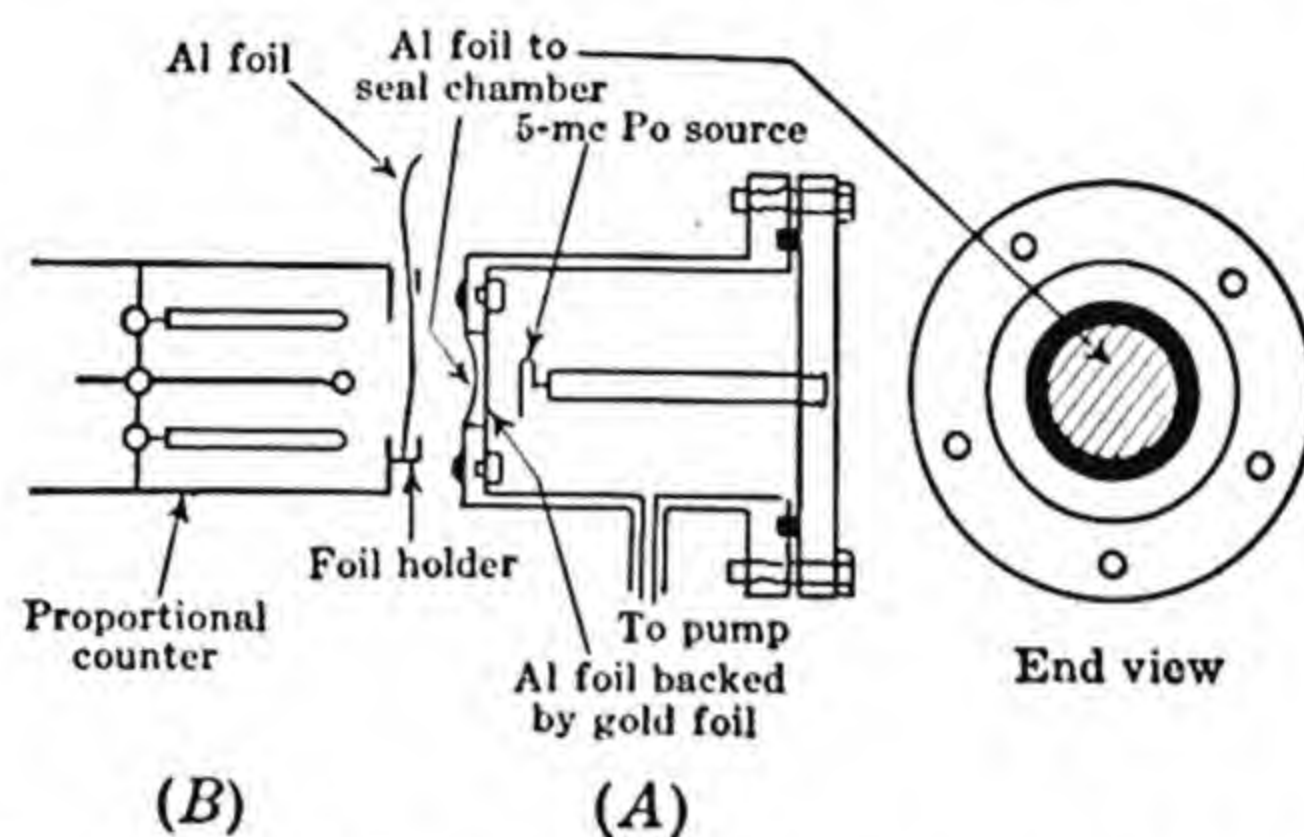
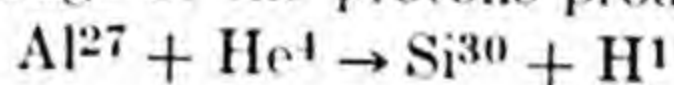


FIG. 1. Bombardment chamber, and proportional counter for alpha-particle transmutation of aluminum. The Al foil at (A) is bombarded by alpha particles from the source. These are stopped in a gold foil placed behind the Al. The chamber is made vacuumtight with an Al foil waxed on to the end. The source is on a holder which is sealed in place with a rubber gasket. Al foils can be placed at (B) to measure the range of the protons produced in the reaction:



source of alpha particles, of radium and beryllium for neutrons (10 millicuries), a source of  $\text{Co}^{60}$  wire for gamma rays and some  $\text{P}^{32}$  for beta radiation. The two former can be obtained from the Canadian Uranium and Radium Corporation or the Eldorado Corporation, and the two latter from Oak Ridge. The  $\text{P}^{32}$  will need to be re-ordered as its half-life is short, but it is excellently handled by the Isotopes Division of the Atomic Energy Commission, and the ordering process is easy and simple.

For detection we suggest a Lauritsen electroscope, a Geiger counter and scaling circuit, and a proportional counter with amplifier and accessories. These can all be purchased on the market. An ingenious circuit man can so easily construct the latter two (including the proportional counter) that we suggest that he amuse himself by building them.



We suggest four experiments, though many more can be devised with this much equipment. The first is a repetition of Rutherford's original transmutation experiment, with the exception that aluminum is studied in place of nitrogen. The apparatus consists of a proportional counter with a foil of 5 cm air equivalent sealing it off and tank argon at about 20-cm pressure for gas filling. A series of aluminum foils (obtainable from the Aluminum Company of America) are cut and weighed and expressed as air equivalent by using the factor  $1.54 \text{ mg/cm}^2$  is equivalent to 1 cm air.

A bombardment chamber is then constructed as indicated in Fig. 1, in which the source bombards a thin aluminum foil placed across an opening which is

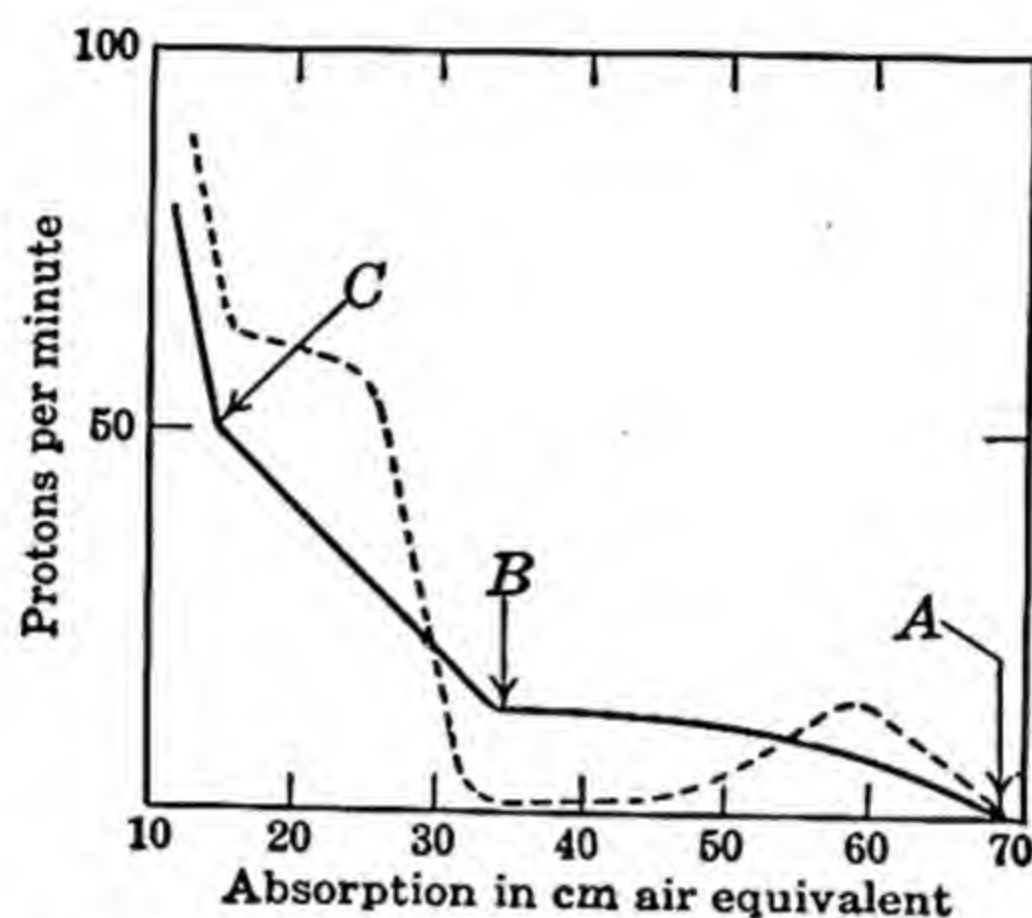


FIG. 2. Absorption curve for protons taken with the bombardment chamber of the previous figure. The dotted curve is for a high bias on the counting circuit. The existence of three groups is seen. At *A* the ground state of  $\text{Si}^{30}$  is formed and the nuclear energy change checks the mass values. At *B* the first excited state of  $\text{Si}^{30}$  is formed, and at *C* are protons produced by bat-and-ball collisions between hydrogen gas contamination in the aluminum and the alpha particles.

further sealed by a gold foil of air equivalent (based on  $3.42 \text{ mg/cm}^2 \equiv 1 \text{ cm air}$ ) of 4 cm. A small foil holder is fastened on to the bombardment chamber which is then evacuated through a glass wool filter to catch any radioactivity due to polonium detaching itself from the source.

The source is placed as shown. It should preferably be deposited on one side of a nickel button, and a 5 millicurie source is ample.

On placing the counter near the end of the bombardment chamber a yield of about 60 counts per minute due to the reaction  $\text{Al}^{27}(\alpha p)\text{Si}^{30}$  should be observed. By interposing aluminum absorbers an absorption curve for the protons should be obtained which will contain three groups. One group, with a total range of about 16 cm, is due to bat-and-ball collisions between alpha particles and hydrogen contamination in the aluminum. The other two are due to ground and first excited states of  $\text{Si}^{30}$ . Measurement of their energies, using the equation of Appendix 7 and the range-energy relation, gives two  $Q$  values. The greatest of these can be shown to check with the mass difference for the reaction, using the masses in the table and the relation  $E = mc^2$  in the correct units.



The kind of absorption curve obtained is shown in Fig. 2. If the bias on the counting circuit is raised, then the dotted appearance results. This is due to the higher rate of ionization at the end of the particle range.

A second experiment is the manufacture of short-lived radiosilver and a measurement of its half-life. For this 10 millicuries of a mixture of radium and beryllium are needed. This is surrounded by a cylinder of paraffin 9 inches in diameter with a 1-inch hole bored in the center. A silver tube  $\frac{3}{4}$  by 4 inches is lowered over the source for 30 seconds and is quickly removed to another

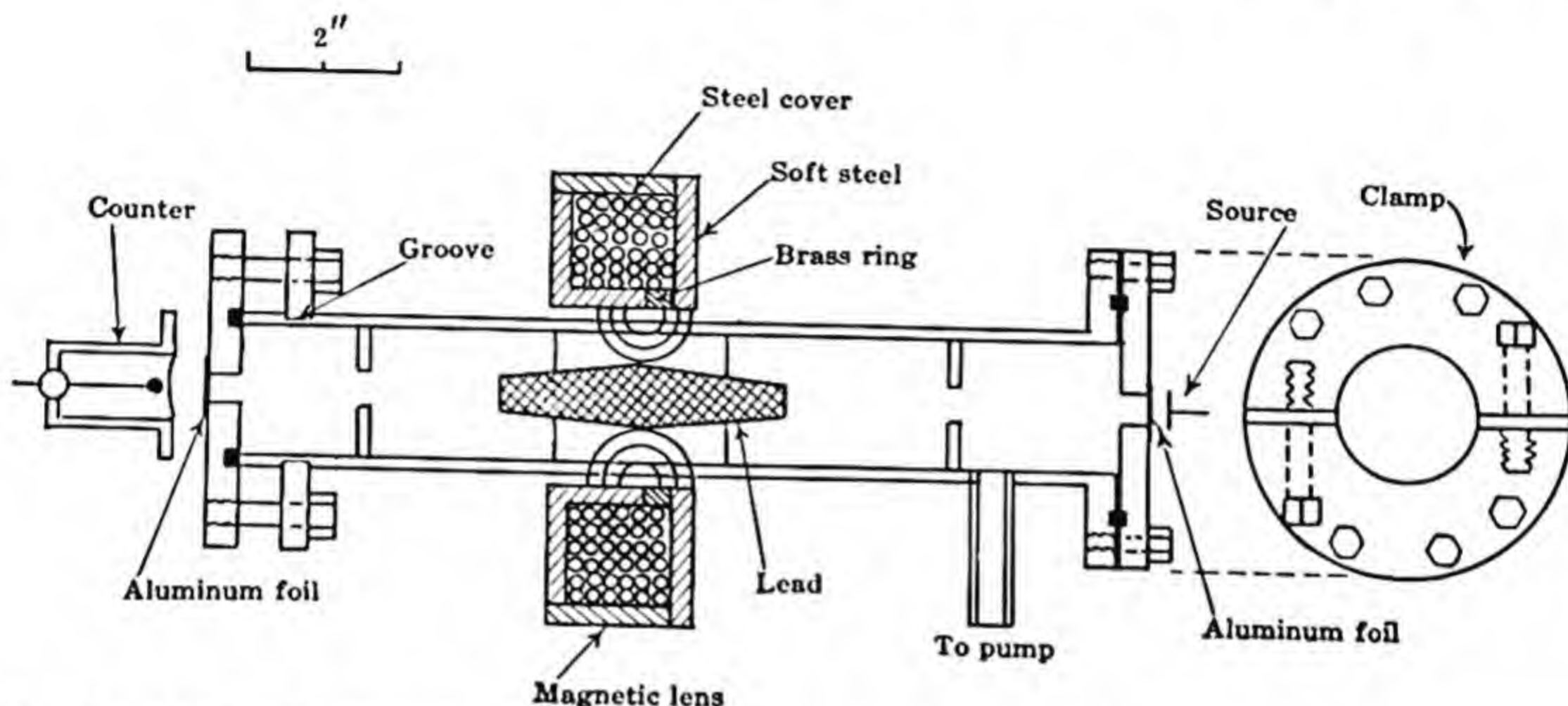


FIG. 3. A simple lens spectrometer. About 1100 turns of No. 16 or No. 18 enamel wire are wound in a steel spool with a gap about  $\frac{3}{8}$  inch wide of brass in the center cylinder. This brass should be welded to steel about  $\frac{1}{4}$  inch thick. The magnetic circuit is closed with a steel cover as shown. The defining slits are two circular holes and a lead double cone. To enable removing the lens the left-hand end has a gasket held by a brass clamp which clamps into a groove as shown. The clamp is made in two sections and so can be removed. A pressure of around 1 mm Hg is necessary.

room (where the background from the 10 millicuries of radium is not observed) and slid over a cylinder Geiger counter. There should result about 100 counts per minute which should decay with a half-life of 22 seconds. This is due to  $\text{Ag}^{110}$  by the reaction  $\text{Ag}^{109}(n\gamma)\text{Ag}^{110}$ . It is likely that some activity due to  $\text{Ag}^{108}$  due to  $\text{Ag}^{107}(n\gamma)\text{Ag}^{108}$  with a half-life 2.3 minutes will be observed. This can be deliberately intensified by exposing the silver to the source for 5 minutes. About 30 counts per minute should result.

The process of sorting out half-lives can be applied to these two periods. Counts should be taken over 6-second intervals, and it will be necessary to expose the silver several times. Ten minutes should be allowed between each exposure, and identical exposure conditions should be used.

A variant on this is to lower the source into a bath of potassium permanganate and to collect the 2.5-hour activity of  $\text{Mn}^{56}$  after an hour's exposure or so. Of course, the source must be in a watertight container, not of Pyrex which contains boron.



Still another variant is to collect 25-minute iodine from ethyliodide by adding a little free iodine, placing the ethyliodide near the source but surrounded by paraffin, and removing the free iodine with KI after exposure. This is the Szilard-Chalmers process and the radioactivity is primarily collected in the free iodine.

A third experiment involves plotting the beta-ray spectrum of  $P^{32}$ . For this a simple lens spectrometer can be constructed, as indicated in Fig. 3. The lens is easy to wind, and the whole spectrometer can readily be built by anyone mechanically inclined. The source of  $P^{32}$  is placed at one end and a bell-

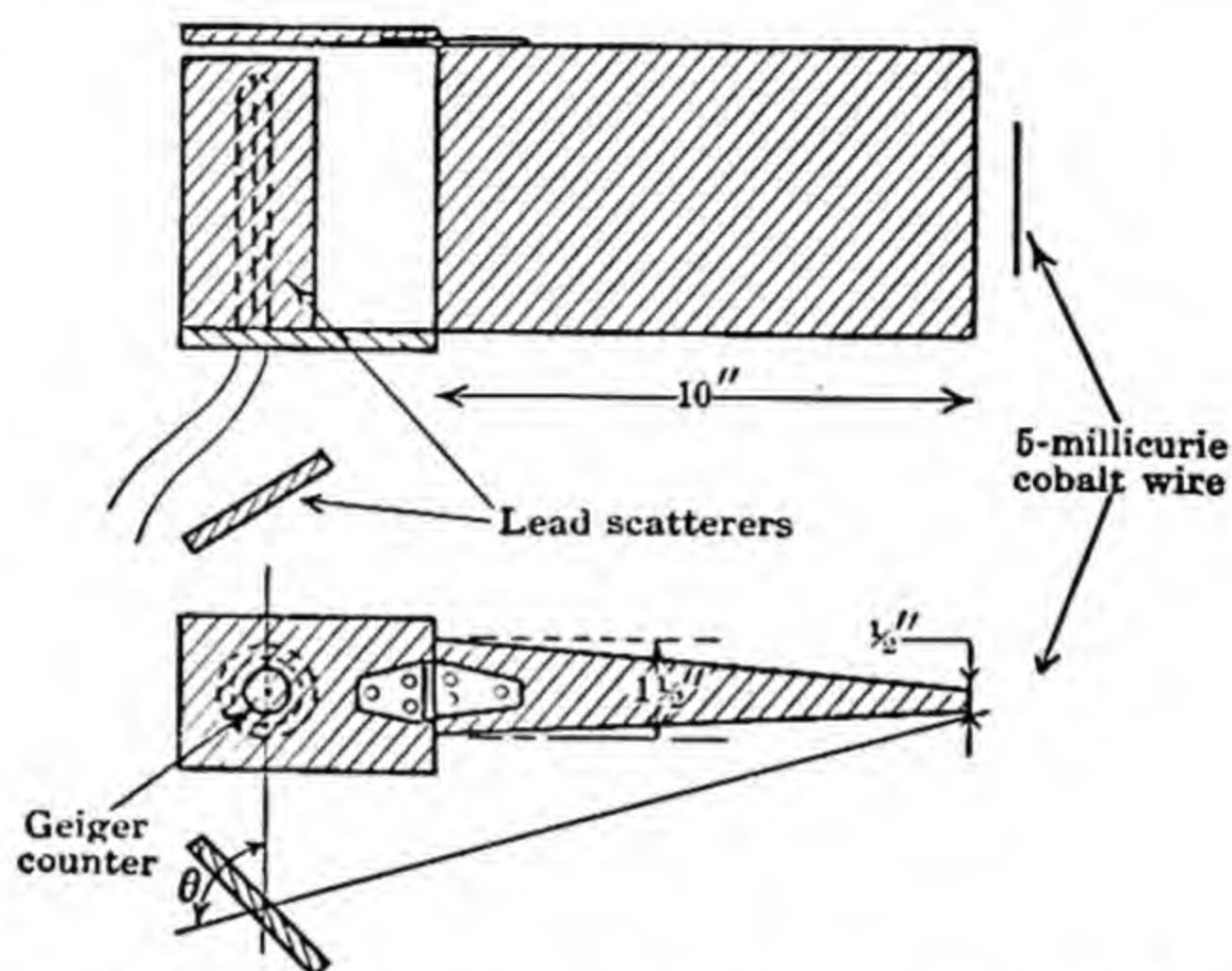


FIG. 4. Apparatus to demonstrate the change of wavelength of gamma rays with scattering. A gamma-ray source of cobalt in the form of wire is shielded directly from a counter by 10 inches of lead. Scatterers of lead as shown are then placed at various positions so as to give various scattering angles. The gamma-ray energy (and hence wavelength) is measured by means of cylindrical absorbers placed around the counter.

shaped beta-ray counter at the other. The current through the coil is varied and a beta-ray spectrum is plotted as indicated. The actual value of  $H\rho$  has to be found by calibration. This is not easy to do, and if a more elaborate experiment is needed it is suggested that  $P^{32}$  be used as a calibration and, say,  $Na^{24}$  be checked against it. Just plotting the beta-ray spectrum alone is usually most interesting.

Low-energy spectra cannot be easily measured because very thin foils are necessary.

As a fourth experiment a check of Compton scattering can be made. For this a  $Co^{60}$  source in the form of wire is placed behind a wedge-shaped lead block about 6 inches high and 10 inches long, tapering to about  $\frac{1}{4}$  inch at the narrow end and  $1\frac{1}{4}$  inch at the wide. Lead is easy to cast.

A cylindrical counter is then arranged as shown in Fig. 4, so that no direct radiation from the source reaches the counter. Lead covers are needed above and below the cylindrical counter. Lead scatterers,  $\frac{1}{16}$  inch thick and about 6 by 6 inches are then placed at various angles, and the absorption curve for the scattered radiation is plotted with cylindrical absorbers placed over the counter.



It will be found that the scattered radiation at high angles is far less penetrating than the direct, and by measuring a series of absorption coefficients the Compton scattering relation can readily be checked. This is

$$\Delta\lambda = \frac{h}{mc} (1 - \cos \theta)$$

where  $\Delta\lambda$  is the wavelength change. Since  $\lambda = c/\nu$  and  $h\nu = E$  where  $\nu$  is frequency and  $E$  is energy, we have

$$\Delta\lambda = \frac{cn}{E^2} \Delta E$$

$\Delta E$  can be figured from the change in absorption coefficient and  $E$  for  $\text{Co}^{60}$  is 1.2 Mev average. The check is remarkably good and this experiment is a good way to convey conviction about the need for a quantum theory. It was first described by J. A. Grey in 1912.

We have taken the attitude that simple experiments such as alpha-, beta-, and gamma-ray absorption need no description on our part and they should be done before any of these are described. For illustrating biological action  $\text{P}^{32}$  and a tomato plant are an excellent combination. The uptake is rapid, and all kinds of measurements, including radioautographs, can easily be made.

A catalog of instruments has been put out by the Radiation Instrument Branch of the Atomic Energy Commission and is available at a low price. This is an excellent guide to the commercially available equipment and should be consulted by anyone intending to set up a laboratory.

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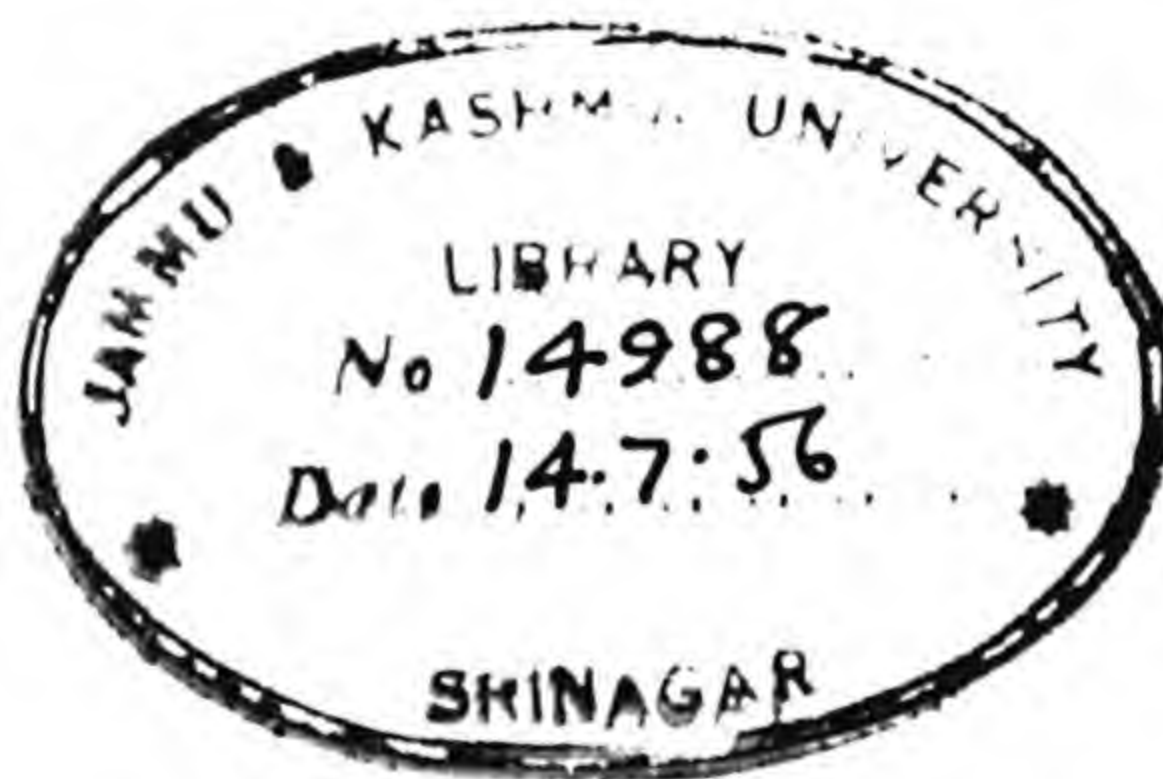
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